Stability of the self-trapped exciton magnetic polaron in diluted magnetic semiconductors: $Cd_{1-x}Mn_xTe$

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The self-trapped exciton magnetic polaron (ST-EXMP) in diluted magnetic semiconductors (DMS's) has been the subject of intensive investigations related to the observed $L2$ photoluminescence during the past two decades, although the stability seems still in controversy. In this paper we have studied the stability condition for ST-EXMP theoretically: we applied the molecular-field approximation to begin with, and then incorporated the effects of the thermodynamic fluctuations of magnetization. The calculation suggests that the exciton magnetic polaron (EXMP) in $Cd_{1-x}Mn_xTe$ ($x=0.05-0.2$) is not self-trapped except at quite low temperatures below ~1 K, but the other primary localization mechanisms, for example, alloy potential fluctuations, are necessary for the localization at the temperatures above ~1 K. This conclusion is different from a recent theoretical report [J. Appl. Phys. **81**, 6297 (1997)] that the ST-EXMP in DMS's is stable up to relatively high temperatures ($T \leqslant 30$ K) over a wide range of Mn concentrations without any other primary localization. The reason for this difference is clarified. It is also discussed briefly that when the higher-order *sp-d* exchange interaction is taken into account, the ST-EXMP in DMS's may become more unstable than that studied in the present paper.

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I. INTRODUCTION

In the last few years, a large number of publications have been devoted to study the physics of diluted magnetic semiconductors (DMS's),[1] in which a part of cations is randomly replaced by transition-metal ions with localized magnetic moments such as the ternary alloy $Cd_{1-x}Mn_xTe$. The local magnetic moments of Mn^{2+} ions strongly interact with the electrons or holes in the conduction and valence bands, producing a variety of substantial phenomena in optical processes, such as the giant Zeeman splitting,[2] the large Faraday rotation,[3] and the formation of so-called exciton magnetic polarons (EXMP's).[4] Among them, the possibility of the photoinduced self-trapped exciton magnetic polaron (ST-EXMP), particularly in $Cd_{1-x}Mn_xTe$, has attracted significant interest related to the observed $L2$ photoluminescence line during the last two decades;[4–11] the $L2$ line shows the Gaussian shape in low temperatures and thus suggests a localization of the exciton. In 1983, Golnik, Ginter, and Gaj[4] discussed the following possible mechanisms: one is the localization by alloy potential fluctuations (APF's), inherent to the mixed crystals, and the other is a magnetic localization of excitons. Even in the latter case, two mechanisms have been suggested: the localization by thermodynamic fluctuations of magnetization and the localization as ST-EXMP's.

Previously, the concept of the magnetic polaron was developed through the study of the optical, magnetic, and transport properties in Eu chalcogeneides (EuX, X=O, S, Se, and Te).[12–17] The stability of the self-trapped magnetic polaron (ST-MP) in EuX, in which the excess carrier is a $5d$ conduction electron and the localized spins arise from $4f$ electrons, has been also investigated; the revealed stability condition, however, is rather severe. In ferromagnetic semiconductors such as EuO and EuS, the ST-MP may be stable at or very near the Curie temperature since the susceptibility of the ferromagnetic material diverges there; however, it can exist for only several degrees around the Curie temperature.[13] For a metamagnetic semiconductor such as EuSe, or an antiferromagnetic semiconductor such as EuTe, the possibility arises that the ST-MP can stably exist at the vicinity of the absolute zero temperature. In the case of EuSe, there is the hope for existence due to the unique characteristics of the interaction between the localized $4f$ spins.[14] In EuTe, however, there seems little possibility.[15,16] Up to now, there has been no conclusive proof for the existence of the ST-MP in EuX from experimental studies. On the other hand, the photoinduced localized magnetic polarons in EuX were realized.[17–19] Recently, the temperature dependence of the spatial extent of the localized magnetic polaron for EuS, EuSe, and EuTe has been deduced[20] from an analysis of the luminescence data.[18]

Let us now return to DMS's. In DMS's, several authors studied the possibility of the ST-MP by the valence hole theoretically using the molecular-field approximation (MFA), and reported that the ST-MP in $Cd_{1-x}Mn_xTe$ can exist only at temperatures below about 1–3 K.[21,22] This may exclude the existence of the ST-EXMP above 1–3 K since the exchange interaction between the conduction electron and localized spins is relatively weak. On the experimental side, by the transient luminescence technique, the time evolution of the exciton localization has been studied; the EXMP localization energy can, however, hardly be distinguished from the energy by APF's, since both contribute to the localization with the same time scale. Takeyama *et al.*,[10,11] however, took up the challenge to separate both contributions; they found that the energy shift of the $L2$ peak above 20 K is insensitive to the temperature, while below 20 K the shift increases drastically with deceasing temperature for the sample with $x=0.12$. They considered the temperature-independent shift above 20 K as a contribution from APF localization, and the temperature-sensitive shift below 20 K as a localization of

the EXMP. Furthermore, comparing with the magnitude of both localization energies, they suggested that the APF's are not necessarily the primary mechanism for the localization of the EXMP. Up to that time, theoretical studies by MFA gave negative answers for the existence of the ST-MP or ST-EXMP at such relatively high temperatures as mentioned above. Then, taking account of the thermodynamic fluctuations of magnetization as well as the attractive Coulomb interaction between the electron and the hole, Miao, Stirner, and Hagston[23] first studied the self-trapping of the EXMP theoretically by employing reasonable values for the material parameters: they found that the ST-EXMP in $Cd_{1-x}Mn_xTe$ is stable up to relatively high temperatures ($T \leqslant 30$ K) over a wide range of Mn concentrations without any other primary localization, e.g., APF's. They demonstrated that the calculated features are consistent with the experimental data reported by Takeyama *et al.*[10,11] Then, in that case, the question arises about what makes the ST-EXMP in DMS's so stabilized.

In this paper, we also study the stability of the ST-EXMP in DMS's, taking account of the thermodynamic fluctuations of magnetization under these circumstances. We will show that the ST-EXMP in $Cd_{1-x}Mn_xTe$ ($x=0.05-0.2$) is rarely realized except at quite low temperatures below ~1 K, but the other primary localization mechanisms, for example, APF's, are necessary for the localization above ~1 K, contrary to the findings of Miao, Stirner, and Hagston.[23] The present paper is organized as follows. The model and the calculated results are given in Sec. II: The molecular-field calculation is performed in Sec. II A, and the thermodynamic fluctuations of magnetization are incorporated in Sec. II B. The conclusion and the several discussions for the stability of the ST-EXMP are given in Sec. III on the basis of the calculation in Sec. II. We also discuss why the conclusion for the stability obtained by the present work is different from that by Miao, Stirner, and Hagston. A brief report of this study has been published in Ref. 24.

II. MODEL AND CALCULATION

Let us consider an electron in the conduction bands and a hole in the valence bands interacting each other through the attractive Coulomb force and also interacting with localized Mn spins embedded in Cd sites by the *sp-d* exchange interaction. Then we start from the following model Hamiltonian,[23]

$$H=-\frac{\hbar^2}{2m_e^*}\nabla_e^2-\frac{\hbar^2}{2m_h^*}\nabla_h^2-\frac{e^2}{\varepsilon|\mathbf{r}_e-\mathbf{r}_h|}$$

$$-I_e\sum_i(\boldsymbol{\sigma}_e)_i\mathbf{S}_i p_i-I_h\sum_i(\boldsymbol{\sigma}_h)_i\mathbf{S}_i p_i, \qquad (2.1)$$

where m_e^* (m_h^*) is the effective mass for electron (hole) in the conduction (valence) bands, $\mathbf{r}_e$ ($\mathbf{r}_h$) is the position vector for electron (hole), ε is the dielectric constant of the crystal, and I_e (I_h) is the constant for the $s(p)$-d exchange interaction defined by

$$I_e=\tfrac{1}{2}N_0\alpha, \quad I_h=\tfrac{1}{2}N_0\beta \qquad (2.2)$$

with the number of cations per unit volume N_0. Furthermore, α (β) is the coupling constant for the exchange interaction between electron (hole) and localized Mn spins, and $\boldsymbol{\sigma}_e$ ($\boldsymbol{\sigma}_h$) is the operator for the electron (hole) spin. The localized Mn spin $\mathbf{S}_i$ is produced by substitution of the Cd ion by a Mn ion: $p_i=1.0$ when the site i is occupied by the Mn ion with $S=\frac{5}{2}$; otherwise $p_i=0.0$. In the ST-EXMP, the localized Mn spins within the EXMP are highly polarized by the *sp-d* exchange interaction and the microscopic ferromagnetic cluster may be established in the paramagnetic medium; the exciton can be self-trapped in this self-induced potential well of the ferromagnetic cluster under certain conditions. Whether such a state stably occurs or not in DMS's is the main subject of the present paper.

In DMS's, the electron (or hole) spin is assumed to be directed in one direction within the spatial extent of the EXMP, because of a small number of the magnetic ions in addition to no magnetic ordering. Thus, in calculating the expectation value of the *sp-d* exchange interaction in Eq. (2.1), the part of the direction of the electron (hole) spin can be separable from the spatial integration as below through neglecting the spatial disorder of Mn ions,[25–27]

$$\langle H_{\rm ex}\rangle=-\boldsymbol{\sigma}_e\cdot I_e x_{\rm eff}\int\int|\phi(\mathbf{r}_e,\mathbf{r}_h)|^2\mathbf{S}(\mathbf{r}_e)d\mathbf{r}_e d\mathbf{r}_h$$

$$-\boldsymbol{\sigma}_h\cdot I_h x_{\rm eff}\int\int|\phi(\mathbf{r}_e,\mathbf{r}_h)|^2\mathbf{S}(\mathbf{r}_h)d\mathbf{r}_h d\mathbf{r}_e \qquad (2.3a)$$

$$=-\boldsymbol{\sigma}_e\cdot\boldsymbol{\Delta}_e-\boldsymbol{\sigma}_h\cdot\boldsymbol{\Delta}_h, \qquad (2.3b)$$

where $x_{\rm eff}$ is the effective Mn concentration explained in Appendix A, and $\boldsymbol{\Delta}_e$ and $\boldsymbol{\Delta}_h$, defined by

$$\boldsymbol{\Delta}_e=I_e x_{\rm eff}\int\int|\phi(\mathbf{r}_e,\mathbf{r}_h)|^2\mathbf{S}(\mathbf{r}_e)d\mathbf{r}_e d\mathbf{r}_h, \qquad (2.4a)$$

$$\boldsymbol{\Delta}_h=I_h x_{\rm eff}\int\int|\phi(\mathbf{r}_e,\mathbf{r}_h)|^2\mathbf{S}(\mathbf{r}_h)d\mathbf{r}_h d\mathbf{r}_e, \qquad (2.4b)$$

are, respectively, so called the spin-splitting vector for the electron and hole.

In the usual MFA, the electron or hole spin is approximated to interact with the thermal average of the each spin-splitting vector: $\langle\boldsymbol{\Delta}_e\rangle_T$ or $\langle\boldsymbol{\Delta}_h\rangle_T$. However, since the localized Mn spins are strongly fluctuated even at low temperatures in DMS's and the number of Mn ions within the EXMP is finite, the deviation of $\boldsymbol{\Delta}_e$ or $\boldsymbol{\Delta}_h$ from each thermal average becomes large for both the direction and the magnitude; the effect of the thermodynamic fluctuations may become relatively large, particularly at high temperatures.

In this paper, then, we first study by the usual MFA and then incorporate the effect of the thermodynamic fluctuations of magnetization. Miao, Stirner, and Hagston[23] took into account the fluctuations of magnetization using the method proposed by Warnock and Wolff[26] (WW); thus, they considered only the *p-d* exchange interaction between the hole and the Mn spins, since the WW method is applicable to only one carrier system. In the present paper, the thermodynamic fluc-

tuations of magnetization are taken into account by extending the fluctuating molecular-field method proposed for the bound magnetic polaron (BMP)[27] to the case of the EXMP. Thus the fluctuations are considered for both spin freedoms of electron and hole.

The following approximations are adopted: (i) the effect of APF's is not taken into account, since the localization due to the purely magnetic origin is studied in this paper. (ii) For the reference system to ST-EXMP, which is another solution of Hamiltonian Eq. (2.1), the free-exciton state, where the exciton is in motion through the crystal and the localized Mn spins are randomly fluctuated, is considered. (iii) For ST-EXMP, the electron-hole pair type, which is a Hartree-type wave function

$$\phi(\mathbf{r}_e,\mathbf{r}_h)=\varphi_e(\mathbf{r}_e)\quad\varphi_h(\mathbf{r}_h) \tag{2.5}$$

is used, where $\varphi_e(\mathbf{r}_e)$ is assumed to be the $1s$ hydrogen-type wave function with the effective Bohr radius a_e:

$$\varphi_e(\mathbf{r}_e)=\frac{1}{\sqrt{\pi a_e^3}}\exp\left(-\frac{r_e}{a_e}\right), \tag{2.6}$$

and $\varphi_h(\mathbf{r}_h)$ is the same type of the wave function with a_h.

With these approximations, the following two cases for a Mn concentration of $x=0.1$ are mainly studied: (a) $m_h^*/m_e=1.25$, $N_0\beta=-1000$ meV. (b) $m_h^*/m_e=0.81$,[28] $N_0\beta=-880$ meV.[29] Here m_e is the free-electron mass. For other material parameters, the following experimental values are employed: $m_e^*/m_e=0.096$,[28] $N_0\alpha=220$ meV,[29] and $\varepsilon=10.6$ (Ref. 30); furthermore, $x_{\mathrm{eff}}=0.039$ and $\Theta=3.84$ K for $x=0.1$.[29] Θ is the phenomenological fitting parameter for an interaction between Mn ions as mentioned in Appendix A; so the case for $\Theta=0$ K is also studied for comparison. The values of the parameters for the valence hole in case (a) are larger than those in case (b) obtained by the experiment. Then, case (a) is favorable to ST-EXMP than case (b), while the case (b) may be closely relevant to $Cd_{1-x}Mn_xTe$. Even in case (b), the (heavy) hole mass is a little bit larger than that usually reported: $m_h^*/m_e=0.6$.[28] This is because the hole mass in case (b) includes the effect of the LO-phonon interaction.[28]

After the calculation of the ST-EXMP, the stabilization free energy is calculated as the difference of the free energy between the ST-EXMP and the free exciton state:

$$F_S=-(F_{\text{ST-EXMP}}-F_{\text{free exciton}}). \tag{2.7}$$

We define the ST-EXMP to be stable compared with free-exciton state when the stabilization free energy, F_S, is positive.

A. Molecular-field approximation

To begin with, we start from the usual MFA. In the MFA, the electron and hole spins interact with the thermal average of each spin-splitting vector, that is, the thermal average of the magnetization of the localized spins $M(\mathbf{r})=\langle S^z(\mathbf{r})\rangle_T/S$, where z is the mean-field axis. Since $\alpha>0$ and $\beta<0$, the mean-field polaron ground state is the doubly degenerate electron-hole spin singlet,[31] which is the state just after the photoexcitation. Then, in the case when the electron spin, $\boldsymbol{\sigma}_e$, is directed to the $+z$ direction, the hole spin, $\boldsymbol{\sigma}_h$, is directed to the $-z$ direction, and vice versa. The free energy for ST-EXMP in this case may be given as

$$\begin{aligned}F_{\text{ST-EXMP}}&(\varphi_e(\mathbf{r}),\varphi_h(\mathbf{r}),M(\mathbf{r}))\\&=-\frac{\hbar^2}{2m_e^*}\int\varphi_e^*(\mathbf{r}_e)\nabla^2\varphi_e(\mathbf{r}_e)d\mathbf{r}_e\\&\quad-\frac{\hbar^2}{2m_h^*}\int\varphi_h^*(\mathbf{r}_h)\nabla^2\varphi_h(\mathbf{r}_h)d\mathbf{r}_h\\&\quad-\frac{e^2}{\varepsilon}\int\int\frac{|\varphi_e(\mathbf{r}_e)|^2|\varphi_h(\mathbf{r}_h)|^2}{|\mathbf{r}_e-\mathbf{r}_h|}d\mathbf{r}_e d\mathbf{r}_h\\&\quad-kT\ln\left\{2\cosh\left(\frac{\Delta_e}{kT}\right)\right\}\\&\quad-kT\ln\left\{2\cosh\left(\frac{\Delta_h}{kT}\right)\right\}-\frac{kT}{V_p}x_{\text{eff}}\int G(M(\mathbf{r}))d\mathbf{r}.\end{aligned} \tag{2.8}$$

In Eq. (2.8), the first and second terms are the localization energies for electron and hole, the third term is the attractive Coulomb interaction energy between electron and hole, the fourth and fifth terms are the free energy due to the *sp-d* exchange interaction for electron and hole, respectively, and the last term is the entropy for the localized spins by MFA. V_p is the volume per cation, k is the Boltzman constant, and T is the temperature. The magnetization $M(\mathbf{r})$ is given by $M(\mathbf{r})=B_S(h(\mathbf{r}))$, where $B_S(h(\mathbf{r}))$ is the Brillouin function with the molecular field $h(\mathbf{r})$ given by

$$\begin{aligned}k(T+\Theta)h(\mathbf{r})&\\&=V_p\left\{I_eS|\varphi_e(\mathbf{r})|^2\tanh\left(\frac{\Delta_e}{kT}\right)+I_hS|\varphi_h(\mathbf{r})|^2\tanh\left(\frac{\Delta_h}{kT}\right)\right\}.\end{aligned} \tag{2.9}$$

Here Θ is the parameter for an interaction between Mn ions determined from experiment (see Appendix A). For the electron-hole spin singlet ST-EXMP, both the molecular fields from the electron and the hole polarize the localized Mn spins cooperatively. Using the variation method, the free energy for ST-EXMP, Eq. (2.8), is minimized with respect to a_e and a_h after $M(\mathbf{r})$ was obtained as a function of a_e and a_h through Eq. (2.9).

On the other hand, the free energy for the free-exciton state is obtained as

$$\begin{aligned}F_{\text{free exciton}}=&-\frac{e^4}{2\varepsilon^2\hbar^2}\left(\frac{1}{m_e^*}+\frac{1}{m_h^*}\right)^{-1}\\&-2kT\ln 2-x_{\text{eff}}N_0kT\ln(2S+1).\end{aligned} \tag{2.10}$$

The first term in Eq. (2.10) is the binding energy due to the relative motion of electron and hole, the second is the entropy for both electron and hole spins, and the third term is the entropy for localized spins. The kinetic energy of the

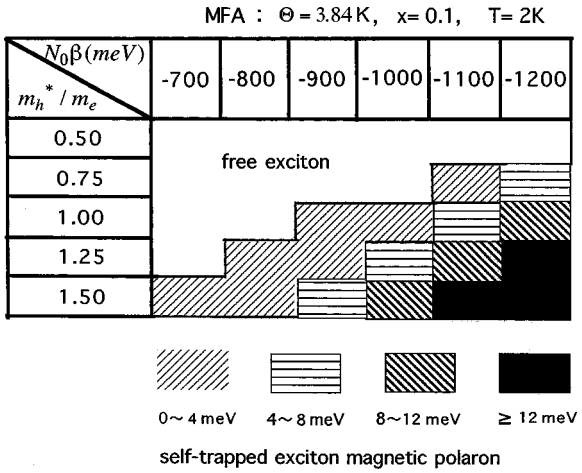

FIG. 1. Map of the stabilization free energy F_S for the ST-EXMP obtained by the MFA is given by varying the hole mass m_h^* and the *p-d* exchange interaction, $N_0\beta$. This is the case for the Mn^{2+} concentration of $x=0.1$, the temperature of 2 K, and $\Theta=3.84$ K. F_S is classified by the blocks with several patterns.

center-of-mass motion is taken as zero since the bottom of the exciton band is considered. The *sp-d* exchange interaction energy for the free-exciton state is zero within the first-order perturbation of the interaction.

We first study the cases (a) and (b) with $\Theta=3.84$ K at 2 K: for comparison, the case with $\Theta=0$ K is also studied. The calculation shows that the stabilization free energy F_S for case (a) with $\Theta=3.84$ K is 6.74 meV, while that for case (b) with $\Theta=3.84$ K is -2.37 meV; the ST-EXMP in case (a) with $\Theta=3.84$ K is, thus, stable at 2 K compared to the free-exciton state, while the free-exciton state is stable at 2 K for case (b) with $\Theta=3.84$ K. To examine the stability for other choices of the material parameters, we perform the same calculation for several sets of m_h^* and $N_0\beta$; the results are shown in Fig. 1 as a map of F_S at $T=2$ K for $\Theta=3.84$ K. It is seen that in the case usually accepted for $Cd_{1-x}Mn_xTe$, $m_h^*/m_e=0.6$ and $N_0\beta=-880$ meV, the ST-EXMP is not stable at 2 K; the ST-EXMP becomes stable when the following approximate inequality is satisfied: $m_h^*/m_e > -0.0017|N_0\beta|+2.5$, where the unit of $N_0\beta$ is taken as meV. The ST-EXMP states calculated at 2 K for cases (a) and (b) are shown in Figs. 2(a) and 2(b), respectively. For case (a) with $\Theta=3.84$ K, the localized Mn spins within ~10 Å from the trapped center are aligned ferromagnetically with almost saturated magnetizations, and furthermore the localized Mn spins situated between 10 and 15 Å are highly polarized; in this ferromagnetic microregion with the substantial polarization of localized Mn spins, the hole wave function is strongly bound with $a_h=14.4$ Å, while the electron wave function is quite loosely bound with $a_e=64$ Å; the self-trapping is, thus, caused by the hole localization due to the strong *p-d* exchange interaction together with the relatively large m_h^*. Similar characteristics are seen for case (b). The ST-EXMP state with $\Theta=0$ K is also shown in Fig. 2 for cases (a) and (b) for comparison; the induced ferromagnetic microregion with almost saturated magnetization considerably enlarges compared with that for $\Theta=3.84$ K, while the

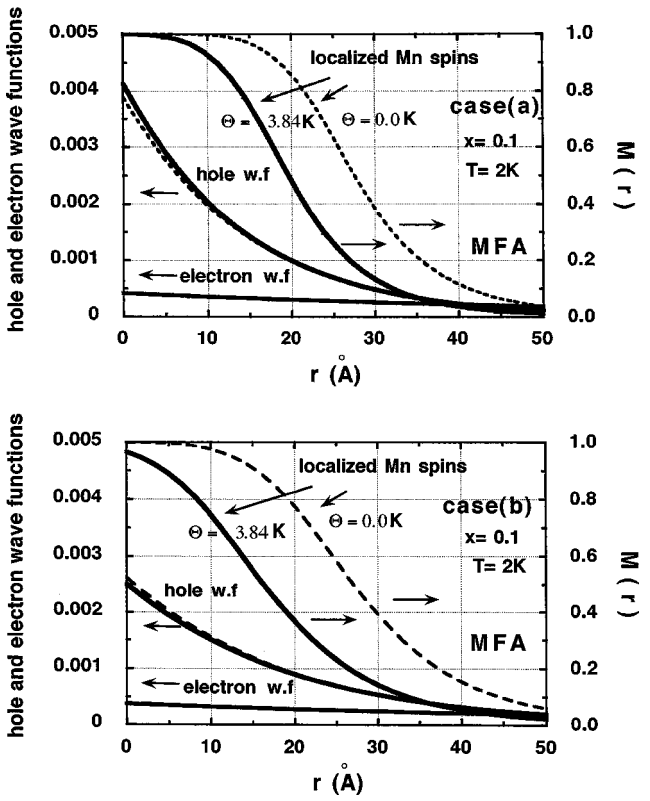

FIG. 2. The state of the ST-EXMP at 2 K calculated by the MFA is shown for $x=0.1$ as a function of the distance from the trapped center. (a) is for case (a), while (b) is for case (b). The hole and electron wave functions are shown with the scale on the left-hand side *y* axis, while the magnetization of the localized Mn spins is shown with the scale on the right-hand side *y* axis. The solid line is for $\Theta=3.84$ K, while the broken line is for $\Theta=0$ K. The figure shows the state within $r=50$ Å; the actual calculation is performed within $r=160$ Å.

extent of the hole and the electron wave function is little changed. Both the ST-EXMP's with $\Theta=0$ and 3.84 K for case (a) shown in Fig. 2(a) are stable. On the other hand, the ST-EXMP for case (b) with $\Theta=0$ K shown in Fig. 2(b) is stable, while that for case (b) with $\Theta=3.84$ K is unstable.

The temperature dependence of the stabilization energy F_S calculated by MFA is shown in Fig. 3 by the thin solid line for $\Theta=0$ K, and by the thin broken line for $\Theta=3.84$ K: we see that the ST-EXMP for case (a) is stable up to $T_c=5.2$ K for $\Theta=3.84$ K and $T_c=6.2$ K for $\Theta=0$ K, while that for case (b) is stable up to $T_c=1$ K for $\Theta=3.84$ K and $T_c=2.4$ K for $\Theta=0$ K. Here, T_c is the critical temperature below which the ST-EXMP is stable. Note that the case for $\Theta=0$ K is purely paramagnetic; in this case the ST-EXMP becomes surely stable as T approaches to zero, since the susceptibility diverges at $T=0$ K.

We further calculate the Mn^{2+} concentration dependence of the stabilization free energy, F_S, for case (a) at 2 K and show in Fig. 4 together with the concentration dependence of the Stokes shift, which is defined by the energy difference between the absorption and the emission. For this calculation, the Mn concentration dependence of both $x_{\rm eff}$ and Θ, discussed in the last paragraph of Appendix A, was used. We

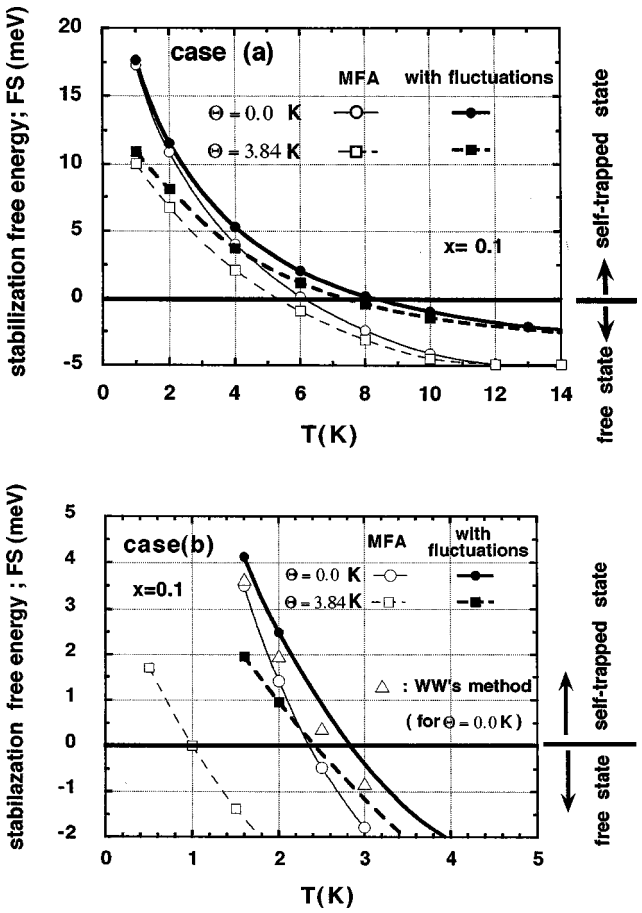

FIG. 3. The temperature dependence of the stabilization free energy F_S is shown for $x=0.1$ for the cases with and without the thermodynamic fluctuations of magnetization. (a) is for case (a), while (b) is for case (b). The result from the MFA is shown by the thin solid or broken line, while that with the fluctuations of magnetization is shown by the thick solid or broken line. The closed or open *circle* is for $\Theta=0$ K, while the closed or open *square* is for $\Theta=3.84$ K. The ST-EXMP is stable for positive F_S. In (b), F_S calculated by the WW method for $\Theta=0$ K is also shown by the mark Δ for comparison.

see that the existence of the minimum concentration of Mn^{2+} ions for the stable ST-EXMP. We also see a consistency between the experiment[10,11] and the calculation for the Mn^{2+} concentration dependence of the Stokes shift. This consistency may suggest the existence of a localized EXMP after the photoabsorption even if the ST-EXMP is not stable.

B. Effect of thermodynamic fluctuations of magnetization

In Sec. II A, we have studied the stability of ST-EXMP by MFA. Judging from the calculation for case (b), which is closely relevant to $Cd_{1-x}Mn_xTe$, we conclude that the ST-EXMP in $Cd_{1-x}Mn_xTe$ at $x=0.1$ may be stable at low temperatures below 1 K for $\Theta=3.84$ K and below 2.4 K for $\Theta=0$ K. This conclusion seems consistent with the previous stability study on the self-trapped hole in DMS's,[21,22] since the *s-d* exchange interaction between the electron and the localized Mn^{2+} spins is fairly weak. The high critical temperature T_c as reported in Ref. 23, namely, 30 K for $Cd_{1-x}Mn_xTe$ or 90 K for $Cd_{1-x}Mn_xSe$, is never obtained by the MFA. Then, does the thermodynamic fluctuations of magnetization drastically improve the stability of ST-EXMP? In Sec. II B, we thus incorporate the effects of the thermodynamic fluctuations of magnetization.

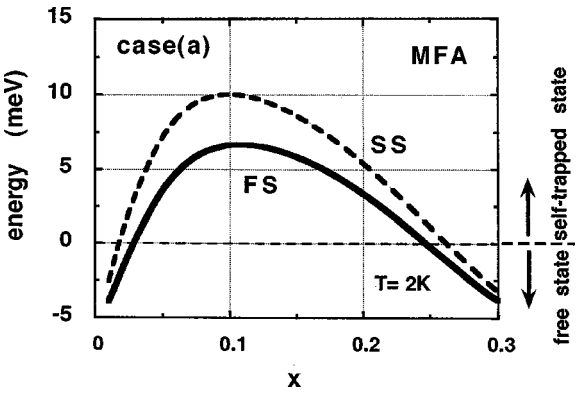

FIG. 4. The Mn concentration dependence of the stabilization free energy F_S calculated by the MFA at 2 K is shown for case (a), together with the Stokes shift (SS). F_S is shown by the solid line, while SS by the broken line.

For the BMP in DMS's, a degree of localized spin alignment around the donor is caused not only by the molecular field produced by the donor electron spin but also by the thermodynamic fluctuation of magnetization.[25–27] This has been clearly confirmed by the comparison between experiment and calculation.[32] Even in the ST-EXMP, the thermodynamic fluctuations of magnetization may assist the localization of EXMP. By extending the fluctuating molecular-field method for the BMP[27] to the case for EXMP, the sum over states[33] for the EXMP with the thermodynamic fluctuations of magnetization may be given by

$$Z=\int\int \exp\left[-\frac{1}{kT}F(T,\mathbf{\Delta}_e,\mathbf{\Delta}_h;[\mathbf{M}(\mathbf{r})])\right]d\mathbf{\Delta}_e d\mathbf{\Delta}_h, \tag{2.11}$$

where $F(T,\mathbf{\Delta}_e,\mathbf{\Delta}_h;[\mathbf{M}(\mathbf{r})])$ is the free energy from the MFA given by Eq. (2.8). $\mathbf{M}(\mathbf{r})$ in Eq. (2.11), however, means one of the possible space profiles of the magnetization for localized Mn^{2+} spins, which produces one of the possible sets of $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_h$ through Eq. (2.4): the thermodynamic fluctuation of $\mathbf{M}(\mathbf{r})$ thus produces the fluctuation of $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_h$. The electron and the hole lower the energy by aligning their spin direction to the direction of instantaneous $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_h$, respectively, but not the direction of the thermal average of $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_h$, since the motion of the electron and the hole spin is fairly faster than that of the localized spins. The integration in Eq. (2.11) should be taken over, in principle, all possible values and directions of the macroscopic order parameter of $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_h$. A sampling method for possible profiles of $\mathbf{M}(\mathbf{r})$ is given in Appendix B. Once Z is obtained, the stabilization free energy for ST-EXMP, F_S, with the thermodynamic fluctuations of magnetization may be given by

$$F_S(a_e, a_h, \xi(T)) = -\left[-kT \ln \int\int \exp\left\{ -\frac{1}{kT} F_{\text{ST-EXMP}}(\mathbf{\Delta}_e, \mathbf{\Delta}_h) \right\} \times d\mathbf{\Delta}_e d\mathbf{\Delta}_h + kT \ln \times \int\int \exp\left\{ -\frac{1}{kT} F_{\text{free exciton}}(\mathbf{\Delta}_e, \mathbf{\Delta}_h) \right\} d\mathbf{\Delta}_e d\mathbf{\Delta}_h \right], \tag{2.12}$$

where $F_{\text{ST-EXMP}}$ is the free energy for EXMP by the MFA for one of the possible sets of $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_h$, and $F_{\text{free exciton}}$ is the free energy for the free-exciton state including the entropy term due to the fluctuation of the localized spins. Furthermore, a_e and a_h are, respectively, the variation parameters for the electron and the hole, and $\xi(T)$ is also a variation parameter[27] associated with the fluctuation of magnetization (see also Appendix B). The trace of the spin for the electron and the hole has been evaluated in $F_{\text{ST-EXMP}}$ and $F_{\text{free exciton}}$, as is shown in Eqs. (2.8) and (2.10). It is worth mentioning that for the free-exciton state, the gain of the *sp-d* exchange interaction by the effect of the thermal fluctuations incorporated in the present study averages out to zero since the free-exciton state spreads over a macroscopic volume.[25,27] We obtain both the ST-EXMP state and F_S with the fluctuations of magnetization by following two methods: one is the method of maximizing F_S, Eq. (2.12), directly with respect to a_e, a_h, and $\xi(T)$. The other is similar to that used in Ref. 27: we obtain a_e, a_h, and $\xi(T)$ by minimizing the free energy of ST-EXMP with the thermodynamic fluctuations of magnetization, namely, by maximizing the first term on the right-hand side of Eq. (2.12), and then calculate Eq. (2.12) with the same a_e, a_h, and $\xi(T)$. Both methods should give the same result in the rigorous calculation. Even in the present approximation, however, we confirmed that both methods give the nearly same result for the temperature range shown in Fig. 3; in this paper, thus, we show the results by the former method. In the actual calculation, we first calculate the case for $\Theta = 0$ K and obtain a_e, a_h, and $\xi(T)$ by the method mentioned above. In the calculation for the case of $\Theta \neq 0$ K, we determine a_e and a_h by maximizing F_S with $\xi(T)$ fixed to the value obtained for the case of $\Theta = 0$ K.

The stabilization free energy F_S calculated in this way is shown in Fig. 3 as a function of temperature: F_S for $\Theta = 0$ K is shown by the thick solid line, while that for $\Theta = 3.84$ K by the thick broken line. F_S obtained by the MFA is also shown for comparison by the thin solid line for $\Theta = 0$ K and by the thin broken line for $\Theta = 3.84$ K. We see from Fig. 3(a) that the stabilization by the thermodynamic fluctuations for case (a) is about 1–2 meV at 2 K and 2.5 meV at 6 K for both cases of $\Theta = 0$ and 3.84 K, as estimated from the energy difference between the thick and thin solid lines for $\Theta = 0$ K, or the thick and thin broken lines for $\Theta = 3.84$ K; the effect of the fluctuations thus stabilizes the ST-EXMP and increases with temperature. By this stabilization, the critical temperature T_c increases: for $\Theta = 0$ K, $T_c = 6.2$ K for the MFA, while $T_c = 8.3$ K when the fluctuations are incorporated. For $\Theta = 3.84$ K, $T_c = 5.2$ K for the MFA, while $T_c = 7.4$ K when the fluctuations are incorporated. The increase of T_c is ~2 K in these cases. Furthermore, F_S for case (b) with the fluctuations of magnetization is shown in Fig. 3(b) in the same way. We see that for $\Theta = 0$ K, $T_c = 2.4$ K for the MFA, while $T_c = 2.8$ K when the fluctuations are incorporated. For $\Theta = 3.84$ K, $T_c = 1.0$ K for the MFA, while $T_c = 2.4$ K with the fluctuations. The increase of T_c is 0.4 K for $\Theta = 0$ K, and 1.4 K for $\Theta = 3.84$ K. As shown by these examples, the effect of the thermodynamic fluctuations of magnetization on T_c is not so large as expected. The reason for this is due to the condition for the stable ST-EXMP that the induced magnetization of localized spins within the polaron should be nearly saturated;[13–16] the energy lowering due to the thermodynamic fluctuations is small for almost saturated regions. From these calculations, we conclude that the ST-EXMP for case (a) is stable below 7.4 K for $\Theta = 3.84$ K and 8.3 K for $\Theta = 0$ K, while that for case (b) is stable below 2.4 K for $\Theta = 3.84$ K and 2.8 K for $\Theta = 0$ K. Above that temperature, the exciton moves freely throughout the crystal without any relaxation of localized Mn^{2+} spins. The high critical temperature T_c as reported in Ref. 23 is not obtained even when the thermodynamic fluctuations of magnetization are incorporated.

III. DISCUSSION AND CONCLUSION

In this section, we make a conclusion for the stability of the ST-EXMP in $Cd_{1-x}Mn_xTe$ and would like to look deeper into the obtained conclusion.

(i) The stability of the ST-EXMP in $Cd_{1-x}Mn_xTe$. As mentioned in Sec. II, the case (b) is closely relevant to $Cd_{1-x}Mn_xTe$. Judging from the calculated result in Sec. II, the ST-EXMP in $Cd_{0.9}Mn_{0.1}Te$ seems unstable except at quite low temperatures below 1–2 K. On the other hand, the transient luminescence experiment in $Cd_{0.88}Mn_{0.12}Te$ certainly shows that the emission energy shift associated with a magnetic localization is observed up to ~25 K, although the energy shift seems continuous with temperature. Provided that the ST-EXMP is stable at low temperatures, it is expected to change to the free-exciton state discontinuously with increasing temperatures.[34] The present calculation together with the experimental result thus suggests that the EXMP in $Cd_{1-x}Mn_xTe$ ($x = 0.05–0.2$) is not self-trapped at temperatures above ~1 K where the experiments have been performed, but is localized by the other primary localization mechanisms, for example, APF's. In this sense, the EXMP in $Cd_{1-x}Mn_xTe$ may resemble the BMP in DMS's, which is primarily localized by the attractive Coulomb interaction between the defect and the electron (hole). The $L2$ luminescence thus is considered to observe the EXMP state trapped by the cooperation of the APF's and the *sp-d* exchange interaction at low temperatures. On the other hand, at high temperatures the luminescence observes the thermodynamic fluctuations of magnetization through the spin freedom of the exciton state trapped by the APF's.

(ii) Comparison with the stabilization energy calculated by the WW method.[26] Since Miao, Stirner, and Hagston[23] applied the method by WW,[26] we also performed the stability

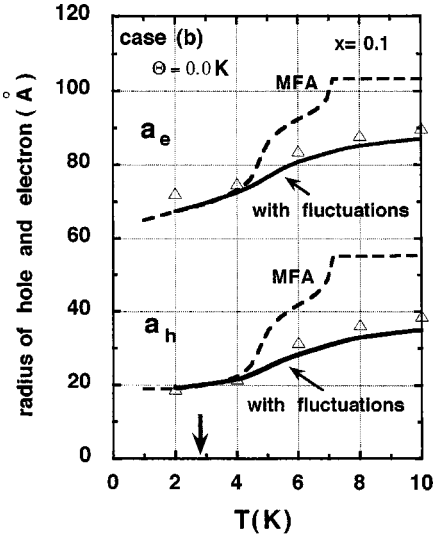

FIG. 5. The extents of the hole and the electron wave function, a_h and a_e, at $x=0.1$ are shown as a function of temperature for case (b) with $\Theta=0$ K. The result by the MFA is shown by the broken line, while that with the thermodynamic fluctuations by the solid line. a_h and a_e obtained by the WW method are also shown by the mark Δ for comparison. The vertical arrow shows T_c below which the ST-EXMP is stable.

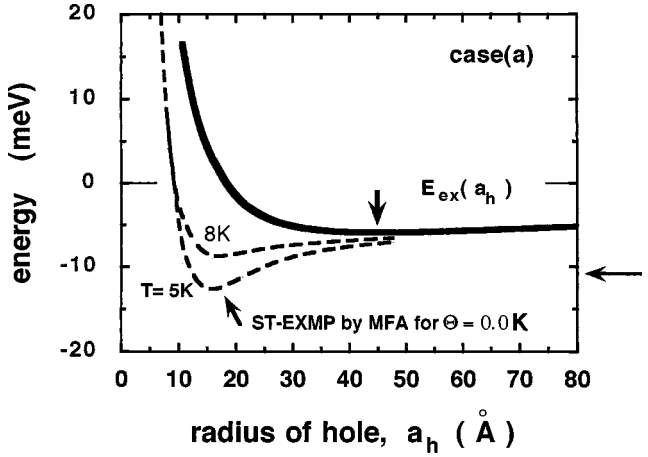

FIG. 6. $E_{\rm ex}(a_h(a_e))$ for case (a) calculated from Eq. (3.1) is shown as a function of the extent of hole a_h by the solid line [see item (iii) in Sec. III]. The vertical arrow shows the minimum of $E_{\rm ex}(a_h(a_e))$, while the horizontal arrow shows the free-exciton energy adopted in the present calculation for case (a). The total free energy for ST-EXMP obtained by the MFA for case (a) with $\Theta=0$ K is plotted as a function of a_h by the broken line for $T=5$ and 8 K, respectively. Note that the free energy for the free-exciton state is obtained by adding $-2kT\ln 2$ (that is, -0.597 meV for $T=5$ K and -0.956 meV for $T=8$ K) to the energy of free-exciton state.

calculation using the same method. The WW method gives an accurate result for the classical localized spins interacting with one excess carrier (that is, only electron or hole) through the exchange interaction; the method however, is applicable only for $\Theta=0$ K. Then, we applied the WW method by taking account of only the exchange interaction between the hole and the localized spins for $\Theta=0$ K, as in Ref. 23. The free energy for ST-EXMP by the WW method is minimized by variation with respect to a_e and a_h. The result obtained for F_S in case (b) is shown by the mark Δ in Fig. 3(b) to be compared with our result shown by the thick solid line [for case (b) with $\Theta=0$ K]; F_S obtained by our calculation is always about 0.5 meV larger than that by the WW method. This seems reasonable since our method takes account of also the exchange interaction between the electron and localized spins: the exchange interaction energy between the electron and localized spins estimated from our calculation is less than 1 meV at temperatures between 1.6 and 4 K. This result therefore may guarantee a validity of the fluctuating molecular-field method used in the present paper. The difference in the stability conclusion between Miao, Stirner, and Hagston[23] and the present work thus is not attributed to the method for calculating the thermodynamic fluctuations of magnetization. The comparison of a_e and a_h between the WW and present method is also shown in Fig. 5, where the mark Δ shows the result by WW, while the solid line shows that by the present method.

(iii) The reason for the difference between Miao, Stirner, and Hagston[23] and the present author. Let us now discuss the reason for the difference in the stability between Ref. 23 and the present work. The independent pair model used for the ST-EXMP, Eq. (2.5), may be valid when the extent of the hole and the electron is small. When the extent becomes quite large, it loses validity since the attractive Coulomb interaction between electron and hole is not taken into account properly. In such a case, it is necessary to describe the wave function as the product of the relative motion and the center of the mass motion for exciton. We therefore estimated the energy of the free exciton on this point of view in the present paper. Nevertheless, Miao, Stirner, and Hagston[23] adopted the independent model even for the free-exciton state as well as ST-EXMP. As a result, they obtained a conclusion different from the present study. To show the situation clearly, we give a brief discussion below. The free-exciton energy in the electron-hole pair model is obtained as a function of a_e and a_h as[23]

$$E_{\rm ex}(a_e,a_h)=\frac{\hbar^2}{2m_h^* a_h^2}+\frac{\hbar^2}{2m_e^* a_e^2}-\frac{e^2(a_h^2+a_e^2+3a_h a_e)}{\varepsilon(a_h+a_e)^3}. \tag{3.1}$$

After minimizing with respect to a_e, the free-exciton energy is only a function of a_h. Then $E_{\rm ex}(a_h(a_e))$ for case (a) is shown as a function of a_h in Fig. 6 by the solid line: the minimum energy is -5.917 meV with $a_h=44.8$ Å. They[23] regarded this minimum energy as the energy for the free-exciton state, while the present study took the first term of Eq. (2.10), -10.79 meV, as the energy of the free-exciton state for case (a). (This energy is indicated by a horizontal arrow in Fig. 6.) Thus, the free-exciton state in the present study is 4.88 meV deeper for case (a), 5.14 meV deeper for case (b) than these cases by them. Their stabilization free energy is always overestimated.

Moreover, when the *sp-d* exchange interaction is turned on, the ST-EXMP for the present approximation appears as

shown by the broken line in Fig. 6. Thus, the free energy for ST-EXMP is always deeper than that for the free-exciton state employed in Ref. 23: the ST-EXMP always stably exists in this point of view. The physical picture of this is as follows. Provided that the free-exciton state is defined as the minimum state of Eq. (3.1), the free-exciton state, that is, the photoexcited state after the interband transition is not extended over the crystal, but is constrained within a finite space characterized by Eq. (3.1). The *sp-d* exchange interaction stabilizes this constrained ''free-exciton state'' and makes it a localized EXMP. In Fig. 5, with increasing temperature, we further see that the extent of the ST-EXMP approaches the extent determined by the minimum of Eq. (3.1). This unphysical result is due to an insufficiency of the pair model. Strictly speaking, thus, the wave function should be described as the product of the relative motion and the center of the mass motion even for ST-EXMP.[34] Further study along this line is now in progress.

(iv) Effects by the second-order sp-d exchange interaction. In the molecular-field approximation in Sec. II A, we used the static approximation that the hole and the electron experience only the average motion of the localized spins, and obtained the free energy of Eq. (2.8). The obtained solution, however, can be taken as the zeroth approximation for the Hamiltonian, Eq. (2.1). We studied the effect of the thermodynamic fluctuations of localized spins in Sec. II B, and showed that the thermal fluctuations make the self-trapped EXMP a little more stable. Another deviation from the static approximation may be estimated by a perturbation calculation of the *sp-d* exchange interaction, which requires the knowledge of the excited state of the magnetic polaron; the calculation of all the excited state is, however, difficult in actual cases. Adopted that the magnetic polaron state is a large polaron as seen in Fig. 2, the excited state may be possibly approximated by the free state orthogonal to the magnetic polaron state obtained by the static model.[16] Then the deviation ΔE_2 from Eq. (2.8) may be estimated as below for the space profile of $M(\mathbf{r})$:

$$\Delta E_2 = -x_{\mathrm{eff}} G \frac{(I_e S)^2}{E_B^e} \int |\varphi_e(\mathbf{r})|^2 [1 - M(\mathbf{r})^2] d\mathbf{r} - x_{\mathrm{eff}} G \frac{(I_h S)^2}{E_B^h} \int |\varphi_h(\mathbf{r})|^2 [1 - M(\mathbf{r})^2] d\mathbf{r}, \quad (3.2)$$

in which E_B^e and E_B^h are, respectively, concerned with the bandwidth of the electron and the hole, and G is a numerical constant of order unity.[16] The proper values of E_B^e, E_B^h, and G for magnetic polarons have not been reported since the difficulties of calculating all the excited states for magnetic polarons. In the free-exciton state, however, Eq. (3.2) tends to the result in the usual second-order perturbation for the *sp-d* exchange interaction by taking $|\varphi_e(\mathbf{r})|^2 = |\varphi_h(\mathbf{r})|^2 = 1/V$ and $M(\mathbf{r}) = 0.0$. Thus, the effect by the second-order perturbation of the interaction is not favorable to the ST-EXMP, since $M(\mathbf{r})$ within the stable ST-EXMP is highly polarized as seen from Fig. 2, while $M(\mathbf{r})$ in the free-exciton state is zero even at the almost absolute zero temperature. The critical temperature T_c thus may descend considerably to the lower temperature than that obtained in Sec. II by the effect of the higher-order perturbation of the exchange interaction.

In this paper, we studied the stability of the self-trapped exciton magnetic polaron in diluted magnetic semiconductors theoretically, related to the observed $L2$ photoluminescence in these substances. First, the usual molecular-field approach was performed, and then the effect of the thermodynamic fluctuations of magnetization was incorporated by the fluctuating molecular-field method. The calculation suggests that the exciton magnetic polaron in $Cd_{1-x}Mn_xTe$ ($x = 0.05-0.2$) is not self-trapped except at quite low temperatures below ~ 1 K, but the other primary localization mechanisms, for example, alloy potential fluctuations, are necessary for the localization at temperatures above ~ 1 K, contrary to the theoretical study by the previous authors.[23] It is briefly discussed further that when the higher-order *sp-d* exchange interaction is taken into consideration, the ST-EXMP in DMS's may become more unstable than that studied in the present paper.

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APPENDIX A

As the concentration of Mn ions increases, the standard Brillouin function becomes inadequate because of the interaction between Mn ions. In such concentration regions, the magnetization is expressed by a modified Brillouin function as[35]

$$M(T,H) = g\mu_B x_{\mathrm{eff}} N_0 S B_S\left(\frac{g\mu_B SH}{k(T+\Theta)}\right), \quad (A1)$$

where N_0 is the number of cations, B_S is the Brillouin function, H is the applied magnetic field, T is the temperature, μ_B is the Bohr magneton, $g=2$, and $S=\frac{5}{2}$ for Mn^{2+} ions. x_{eff} and Θ are fitting parameters to the experimental data and are, respectively, regarded as the effective Mn concentration and an antiferromagnetic temperature. The values for x_{eff} and Θ for $Cd_{0.9}Mn_{0.1}Te$ used in Sec. II were employed from the experimental data.[29] The Mn concentration dependence on x_{eff} has been theoretically proposed also on an isolated-spin cluster model, in which the Mn spins in each cluster interact with others antiferromagnetically, however, there is no interaction among different clusters. An effective Mn concentration x_{eff} on this model was, for example, reported by Shapira *et al.*,[36] considering four types of the nearest-neighbor clusters: isolated Mn ions, pairs, open triangles, and closed tri-

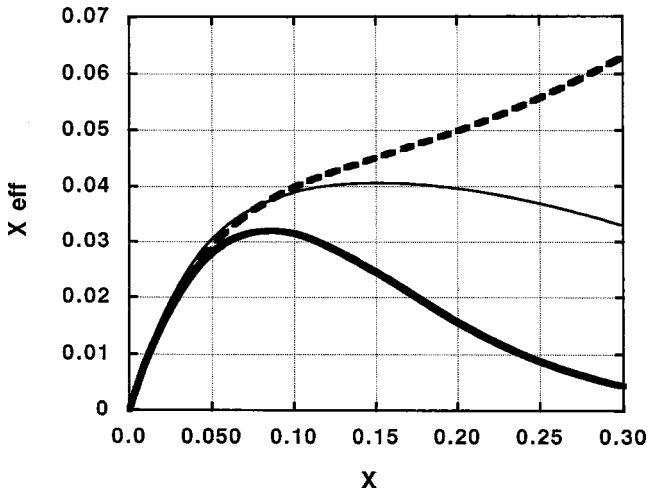

FIG. 7. The effective Mn concentration x_{eff} calculated by Shapira *et al.* (Ref. 36) is shown as a function of x by the thick solid line while the upper bound for x_{eff} by the thick broken line. x_{eff} obtained by a numerical simulation by Fatah *et al.* (Ref. 37) is also shown by the thin solid line (see Appendix A).

angles, together with the upper bound for x_{eff}. The result is shown in Fig. 7, where x_{eff} is shown by the thick solid line and the upper bound for x_{eff} by the thick broken line. Numerical simulation for x_{eff} was also performed by Fatah *et al.*[37] in which the nearest-neighbor unpaired magnetic ions were searched under a random distribution of magnetic atoms. The result is shown by the thin solid line in Fig. 7, which shows an intermediate value between the thick solid and broken lines obtained by Shapira *et al.*[36]

In Sec. II A, we calculated the stabilization free energy and the Stokes shift as a function of the Mn concentration x, which requires the Mn concentration dependence of both x_{eff} and Θ; we used x_{eff} obtained by the numerical simulation by Fatah *et al.*[37] mentioned above, and also used the following antiferromagnetic temperature Θ interpolated for the experimental data[29] between $x=0.005$ and 0.3: $\Theta=56.02x-251.02x^2+766.18x^3$ K. When we restrict the fitting area between $x=0.005$ and 0.2, $\Theta=(37.45\pm1.06)x$ K represents the experimental data[29] well.

APPENDIX B

In this appendix, we show how to generate the thermodynamical fluctuations of magnetization in Sec. II B. In this method, instead of all possible fluctuated states of magnetization, we take account of the fluctuated states that seem to be effective to the problem.[27] We introduce a fluctuating molecular field on the localized spin at $\mathbf{r}$, $\mathbf{H}(\mathbf{r}:\theta_e,\theta_h,\xi_e,\xi_h,\nu_e,\nu_h)$, and divide it into two components:

$$\mathbf{H}(\mathbf{r}:\theta_e,\theta_h,\xi_e,\xi_h,\nu_e,\nu_h)$$
$$=\mathbf{H}_e(\mathbf{r}:\theta_e,\xi_e,\nu_e)+\mathbf{H}_h(\mathbf{r}:\theta_h,\xi_h,\nu_h). \tag{B1}$$

Here the components $\mathbf{H}_e$ and $\mathbf{H}_h$ are assumed to be two independent fluctuating molecular fields resulting from two excess carriers, the electron and the hole, for example; they are fluctuated in both the direction and the magnitude: θ_e (θ_h) is the angle between z axis and $\mathbf{H}_e$ ($\mathbf{H}_h$), and ξ_e (ξ_h) is the angle between the x axis and $\mathbf{H}_e$ ($\mathbf{H}_h$). $\mathbf{H}_e$ and $\mathbf{H}_h$ are further described as

$$\mathbf{H}_i(\mathbf{r}:\theta_i,\xi_i,\nu_i)=R_i(r:\nu_i)\mathbf{e}_i(\theta_i,\xi_i), \tag{B2}$$

with

$$\mathbf{e}_i(\theta_i,\xi_i)=\frac{\mathbf{H}_i(\mathbf{r}:\theta_i,\xi_i,\nu_i)}{|\mathbf{H}_i(\mathbf{r}:\theta_i,\xi_i,\nu_i)|} \quad (i=e \text{ and } h). \tag{B3}$$

Therefore, when θ_e and ξ_e (θ_h and ξ_h) are varied, the direction of $\mathbf{H}_e$ ($\mathbf{H}_h$) is fluctuated; when ν_e (ν_h) is varied, the magnitude of $\mathbf{H}_e$ ($\mathbf{H}_h$) is fluctuated. In the present calculation, we employed the $1s$ hydrogen-type function for R_i $(r:\nu_i)$, which is the same type of function as the variation wave function for electron and hole. Thus, R_i $(r:\nu_i)$ is explicitly shown as

$$R_i(r:\nu_i)=V_p(\tilde{I}_iS)(\nu_i^3/\pi)\exp(-2\nu_i r). \tag{B4}$$

Then, when we take $\nu_e=1/a_e$, $\nu_h=1/a_h$, and further $\theta_e=\theta_h=0$ or $\theta_e-\theta_h=\pi$, together with $\tilde{I}_e=I_e$ and $\tilde{I}_h=I_h$, Eq. (B1) reproduces the usual molecular field as expressed by Eq. (2.9). This is why the present method is called a fluctuating molecular-field method. Now, the magnetization fluctuating at $\mathbf{r}$, $\mathbf{M}(\mathbf{r})$, is given by

$$\mathbf{M}(\mathbf{r}:\theta_e,\theta_h,\xi_e,\xi_h,\nu_e,\nu_h)$$
$$=B_S(|\mathbf{H}(\mathbf{r}:\theta_e,\theta_h,\xi_e,\xi_h,\nu_e,\nu_h)|/\xi(T)k(T+\Theta))\mathbf{e}(r). \tag{B5}$$

In the present approximation, all the magnetization vectors for EXMP exist in the plane spanned by $\mathbf{H}_e$ and $\mathbf{H}_h$; within this plane the direction of magnetization, however, depends on r, as shown by the unit vector $\mathbf{e}(r)$, since two independent $\mathbf{H}_e$ and $\mathbf{H}_h$ are used. This ensures the possibility that the direction of the spin-splitting vector for hole $\mathbf{\Delta}_h$ is different from that for electron $\mathbf{\Delta}_e$ for each instance in the fluctuation. In Eq. (B5), B_S is the Brillouin function, and the $\xi(T)$ is a variation parameter, which is determined by minimizing the free energy with the thermodynamic fluctuations of magnetization.[27] The fluctuating magnetization $\mathbf{M}(r)$ is generated from the fluctuating molecular field in this way.

Now, we can calculate the spin-splitting vectors, $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_h$ for the electron and the hole by Eq. (2.4), and also the free energy for the ST-EXMP $F_{\text{ST-EXMP}}$ by Eq. (2.8) using the *instantaneous magnetization* $\mathbf{M}(r)$ given by Eq. (B5). In this process, both the electron and hole spins are assumed to adiabatically follow to each *instantaneous spin-splitting vector* since the motion of the electron and hole spins is considerably faster than that of localized spins. In the present method, the fluctuation is produced when θ_e, θ_h, ξ_e, ξ_h, ν_e, and ν_h are varied. Then in changing the variables of the integration from $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_h$ to θ_e, θ_h, ξ_e, ξ_h, ν_e, and ν_h in Eq. (2.11), the transformation factor, the so-called Jacobian, is required:

$$J(\nu_e,\nu_h,\cos\theta_e,\cos\theta_h,\xi_e,\xi_h) = \frac{\partial(\Delta_e,\Delta_h,\cos\chi_e,\cos\chi_h,\eta_e,\eta_h)}{\partial(\nu_e,\nu_h,\cos\theta_e,\cos\theta_h,\xi_e,\xi_h)}, \quad \text{(B6)}$$

in which χ_e (χ_h) is the angle between the z axis and $\mathbf{\Delta}_e$ ($\mathbf{\Delta}_h$), while η_e (η_h) is the angle between the x axis and $\mathbf{\Delta}_e$ ($\mathbf{\Delta}_h$). Here, the Jacobian is defined, for example, for the case of two variables as $J(p,q)=\partial(x,y)/\partial(p,q)$, and is given explicitly by

$$J(p,q)=\begin{vmatrix} \frac{\partial x}{\partial p} & \frac{\partial y}{\partial p} \\ \frac{\partial x}{\partial q} & \frac{\partial y}{\partial q} \end{vmatrix}. \quad \text{(B7)}$$

Since

$$d\mathbf{\Delta}_e d\mathbf{\Delta}_h = \Delta_e^2\Delta_h^2 d\Delta_e d\Delta_h d(\cos\chi_e) d(\cos\chi_h) d\eta_e d\eta_h, \quad \text{(B8)}$$

the integration by $d\mathbf{\Delta}_e d\mathbf{\Delta}_h$ in Eq. (2.11) is changed to

$$\Delta_e^2\Delta_h^2 J(\nu_e,\nu_h,\cos\theta_e,\cos\theta_h,\xi_e,\xi_h) d\nu_e d\nu_h \times d(\cos\theta_e) d(\cos\theta_h) d\xi_e d\xi_h. \quad \text{(B9)}$$

When there is no external magnetic field, the z axis is no special axis; the free energy Eq. (2.8) thus depends on the relative angle between $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_h$ together with their magnitudes. Thus, sextuple integrals appeared in Eq. (B9) reduce to triple integrals for the case without the external magnetic field studied in this paper; the integration for ν_e and ν_h is performed from 0 to each upper value numerically; the upper value for ν_e and ν_h was estimated with consulting the ST-EXMP state obtained by the MFA. As for $\tilde{I}_i$ in Eq. (B4), one possible choice is $\tilde{I}_e=I_e$ and $\tilde{I}_h=I_h$; this choice was used for the present study conventionally. Other choices to generate proper fluctuations, such as $\tilde{I}_e=\tilde{I}_h=\frac{1}{2}(|I_e|+|I_h|)$ with the same upper value for ν_e and ν_h, are also possible.

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