Free magnetic polaron in a Van Vleck semimagnetic semiconductor

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The self-localization of a free carrier as the result of a carrier-ion exchange interaction is analyzed for the case of a semimagnetic semiconductor of the type $A_{1-x}Fe_xB^6$, whose magnetic ions have a singlet ground state. A simple model is proposed. It reduces the description of a free magnetic polaron at low temperatures to the solution of a nonlinear differential equation in which magnetic saturation of the medium is taken into account. The basic characteristics of the polaron—its energy, the radius of its state, its effective mass, and its g-factor—are calculated as a function of the concentration of the magnetic component. The effect of an external magnetic field on the polaron is analyzed. Possible experimental manifestations of this effect are discussed.

1. INTRODUCTION

Magnetopolaron effects in semimagnetic semiconductors of the type $A^2_{1-x}Mn_xB^6$, whose magnetic ions have an orientational paramagnetism, are being studied widely. These effects have been seen in the temperature dependence of the field-free shift of the line corresponding to a spin-flip Raman scattering by shallow donors,[1-4] in the red shift of the luminescence line of excitons bound to shallow acceptors,[5,6] and in the magnetic-field dependence of the luminescence energy of excitons in quantum wells based on semimagnetic semiconductors.[7-9] A theory for these effects was derived in Refs. 3 and 10–12. That theory shows that, while the magnetopolaron effects associated with a carrier-ion exchange interaction fall off monotonically with increasing T for weakly bound carriers localized at defects, over a broad temperature range the self-localization of free carriers in three-dimensional and two-dimensional systems[10] is a process involving a threshold.

Recent years have seen active research on semimagnetic semiconductors of another type: the so-called Van Vleck semimagnetic semiconductors, whose paramagnetism is of a polarization nature, i.e., is associated with a mixing of excited magnetic states with the singlet ground state of the ions, induced by a magnetic field $\mathbf{H}$ (Refs. 13–15). The typical semimagnetic semiconductors are solid solutions with the general formula $A^2_{1-x}Fe_xB^6$, in which the $3d^6$ ground state of the spin-orbit multiplet for the Fe^{2+} ions is a nonmagnetic singlet A_1, while the T_1, E, T_2, and A_2 multiplets have a nearly equidistant energy spacing with an interval $\Delta\varepsilon = 1.5$–2 MeV. Near liquid-helium temperatures, essentially all the Fe^{2+} ions are thus in the lowest level, and have a purely paramagnetism polarization. In this situation, there will evidently be features in the polaron effect which will distinguish the effect in this case from that in an orientational semimagnetic semiconductor. These features will stem from the absence of spin fluctuations in Van Vleck semimagnetic semiconductors. This circumstance was discussed in Refs. 16 and 17 in connection with the spectra of the spin-flip Raman scattering by shallow donors in Van Vleck semimagnetic semiconductors and a possible manifestation of a polaron effect in these spectra.[18] Optical studies[13,14] and photoelectric studies[19] of the exciton spectra as well as studies of transport phenomena in Van Vleck semimagnetic semiconductors[20] have ignored the possibility of a self-localization of charge carriers due to the formation of a free magnetic polaron in these semiconductors.

In the present paper we propose a model for a free large-radius magnetic polaron in a Van Vleck semimagnetic semiconductor at low temperatures in the singlet ground state. A feature which distinguishes this model in a fundamental way from, for example, the case of a Pekar polaron[21,22] is the allowance for the nonlinearity stemming from the magnetic saturation of the medium. If this saturation were ignored, the polaron would collapse.[23,24] As a result we derive an energy functional for the electron wave function. The corresponding equation determining the extremum reduces to a nonlinear Schrödinger equation only if this saturation is ignored. We then use a direct variational method to calculate the basic characteristics of the polaron. We use an oscillator wave function as a test wave function, and we use the radius of the polaron state as a variational parameter. In particular, we calculate the magnetic-field dependence of the critical concentration of the magnetic component, i.e., the concentration above which self-localization becomes possible. We also calculate the energy, effective mass, and g-factor of the polaron. Numerical estimates for the Van Vleck semimagnetic semiconductor $Zn_{1-x}Fe_xSe$ show that a possible self-localization of holes must be taken into consideration at $x \geqslant 0.04$.

2. EQUATION FOR THE POLARON WAVE FUNCTION

In deriving an equation for the ground state of a free charge carrier in a Van Vleck semimagnetic semiconductor, we start from the Hamiltonian for an electron (or hole) and for magnetic ions which are exchange-coupled with this electron and which randomly replace N_l cations at the sites $\mathbf{R}_j$ ($j = 1,2,...,N_l$) of a cubic crystal lattice. Under the assumption that the corresponding energy band is isotropic and nondegenerate, we write the Hamiltonian of this system in the effective-mass approximation:

$$\mathscr{H} = \frac{\mathbf{p}^2}{2m^*} + g_e\mu_B\mathbf{H}\mathbf{S}_e + \sum_{j=1}^{N_l}[-J\Omega\delta(\mathbf{r}-\mathbf{R}_j)\mathbf{S}_l{}^j\mathbf{S}_e + \mathscr{H}_l{}^j(g_l\mu_B\mathbf{H})] + \mathscr{H}_{ll}, \quad (1)$$

where $\mathbf{p}$, m^*, g_e, $\mathbf{S}_e$, and $\mathbf{r}$ are the momentum operator, the

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effective mass, the band g-factor, the spin operator, and coordinate of the electron; g_l and $\mathbf{S}_l^j$ are the g-factor and spin operator of the magnetic ion localized at lattice site $\mathbf{R}_j$; J is the constant of the carrier-ion exchange interaction; Ω is the volume of the unit cell; μ_B is the Bohr magneton; and $\mathbf{H}$ is the external magnetic field. The first two terms in expression (1) determine the kinetic energy and the Zeeman energy of an electron. The next two terms determine the carrier-ion exchange interaction and the sum of the Hamiltonians of the magnetic ions in the field $\mathbf{H}$. The latter are conveniently written in the approximation of a mean crystal field:[25]

$$\mathscr{H}_l^j(g_l\mu_B\mathbf{H})=\sum_{kq}B_k^qO_k^q+\lambda\mathbf{L}^j\mathbf{S}_l^j+g_l\mu_B\mathbf{H}\mathbf{S}_l^j, \tag{2}$$

where $O_k^q=O_k^q(\mathbf{L}_j)$ are equivalent operators, B_k^q are the corresponding values of the crystal field (the extent to which they differ from zero and the relations between them are dictated by the symmetry conditions), and the operator $\mathbf{L}^j$ represents the orbital angular momentum of the $3d^n$ electrons. The first term in (2) describes the splitting of the orbital multiplet of the magnetic ion by the crystal field. The second term describes the further splitting by the spin-orbit interaction, with a constant λ. We are considering the case in which the lowest-energy level resulting from this splitting is a nonmagnetic singlet. The third term in (2), which corresponds to the Zeeman energy of the ion, leads to a paramagnetic shift of the lowest level as the result of a mixing of excited magnetic states with the ground state. The last term in (1) describes the spin-spin interactions between magnetic ions; of these interactions, the exchange interaction is dominant.

We adopt several assumptions in order to derive expressions for the energy of a free magnetic polaron from Hamiltonian (1).

1) We restrict the analysis to a Van Vleck semimagnetic semiconductor with a magnetic-ion concentration n_l low enough for the interaction $\mathscr{H}_{ll}$ in (1) can be ignored. We can then seek the total wave function of the system in a multiplicative form:

$$\Psi=\psi_e(\mathbf{r})X_e\prod_{j=1}^{N_l}\psi_l^j, \tag{3}$$

where $\psi_e(\mathbf{r})$ and χ_e are respectively the coordinate part and spin part of the electron wave function, and ψ_l^j is the wave function of magnetic ion j, which is determined by Hamiltonian (2).

2) On the other hand, the concentration of magnetic ions must be high enough to satisfy $n_la^3\gg 1$, where a is the effective radius of the free magnetic polaron. The validity of this condition must be tested later. Under this condition, we can treat the crystal as a continuum, and in the calculation of the polaron energy we can replace sums over magnetic ions by integrals over the volume.

3) In calculating the energy of a free magnetic polaron, we ignore the contribution to the energy of the system from that part of the operator representing the carrier-ion exchange interaction which is not diagonal in the spins, as in the case of orientational semimagnetic semiconductors:

$$\mathscr{H}_{sd}'=-{}^1/_2J\Omega\sum_j\delta(\mathbf{r}-\mathbf{R}_j)(S_{l+}^jS_{e-}+S_{l-}^jS_{e+}). \tag{4}$$

It may, on the other hand, turn out to be important to incorporate this interaction for an excited state of a polaron,[18] if the energy of the latter is close to the splitting $\Delta\varepsilon$ in the spin-orbit multiplet of the magnetic ion. In general, the correction for interaction (4) is inversely proportional to n_la^3, and it is small according to assumption 2. The diagonal term, on the other hand, which has the structure of a Zeeman energy, is combined with the last term in (2). The result is to replace $\mathscr{H}_l^j(g_l\mu_B\mathbf{H})$ by $\mathscr{H}_l^j(g_l\mu_BH_z-J\Omega\delta(\mathbf{r}-\mathbf{R}_j)S_{ez})$. Here the z axis runs parallel to the field $\mathbf{H}$.

4) In analyzing the effects in the magnetic field $\mathbf{H}$, we assume that $\mathbf{H}$ is weak enough that the corresponding magnetic length satisfies $a_H\gg a$, and we can ignore the effect of the diamagnetic corrections on the self-localization of free carriers.

5) Finally, in calculating the energies of the magnetic ions described by Hamiltonian (2), we ignore the insignificant effects of the cubic anisotropy, under the assumption that the paramagnetic susceptibility of the Van Vleck semimagnetic semiconductor is the same as the susceptibility of the magnetic ions calculated for the case $\mathbf{H}\|[001]$, for an arbitrary direction of the magnetic field. The latter susceptibility is found in terms of the energy of the ground state of the magnetic ion:[26]

$$E_1(g_l\mu_BH)=\Delta\varepsilon-[\Delta\varepsilon^2+4(g_l\mu_BH)^2]^{1/2}, \tag{5}$$

where $g_l=2$.

Under these assumptions we can find an energy functional for a free magnetic polaron, defined as the difference between the lowest-lying eigenvalues of Hamiltonian (1) and of the Hamiltonian of a Van Vleck semimagnetic semiconductor in the absence of free carriers. We find this functional in the form

$$E_\sigma=\frac{\hbar^2}{2m^*}\int(\nabla\psi_e)^2dV+g_e\mu_BH\sigma+n_l\times\int[E_1(g_l\mu_BH+A\sigma)-E_1(g_l\mu_BH)]dV, \tag{6}$$

where $\sigma=\pm 1/2$ are the eigenvalues of the operator which projects the electron spin onto $\mathbf{H}$, and $A(\mathbf{r})=-J\Omega|\psi_e|^2$ is the effective exchange field. According to assumption 3, this field is the energy of the exchange interaction of the magnetic ion with the electron.

An important feature of functional (6) is its nonlinear dependence on $|\psi_e|^4$, a consequence of the saturation of magnetic energy (5). As a result, the corresponding equation which minimizes (6) with respect to ψ_e does not reduce to the usual nonlinear Schrödinger equation for the problem of self-localization with a short-range potential.[24] It can describe a self-localization of such a nature that a polaron does not collapse, regardless of the parameter values of the problem.

3. ENERGY AND RADIUS OF THE STATE

To find the principal characteristics of the ground state of the polaron, we minimize functional (6) by a direct variational method. Calculations carried out for several single-parameter trial functions have shown that the best is a function

$$\psi_e=(2/\pi a^2)^{3/4}\exp(-r^2/a^2). \tag{7}$$

Substituting (7) into (6), and using (5), we find the following expression for the energy of a polaron in the case $J>0$, at a fixed value of σ:

$$\varepsilon_\sigma(\rho)=\frac{1}{\rho^2}+\varkappa h\sigma$$

$$-\frac{\rho^3}{u}\int_0^\infty\left\{\left[1+4\left(h-\frac{\sigma}{\rho^3}\exp(-2y^2)\right)^2\right]^{1/2}-(1+4h^2)^{1/2}\right\}y^2\,dy. \tag{8}$$

Here we have introduced the dimensionless quantities

$$\varepsilon_\sigma=\frac{4m^*|J\Omega|^{2/3}E_\sigma}{3\pi\hbar^2(\Delta\varepsilon)^{2/3}},\quad h=\frac{g_l\mu_B H}{\Delta\varepsilon},$$
$$y=\frac{r}{a},\quad \rho=a\left(\frac{\pi}{2}\right)^{1/2}\left(\frac{\Delta\varepsilon}{|J\Omega|}\right)^{1/3}, \tag{9}$$
$$u=\frac{3\hbar^2(2\pi)^{2/3}(\Delta\varepsilon)^{2/3}}{128m^*n_l|J\Omega|^{5/3}},\quad \varkappa=\frac{4g_e}{3\pi g_l\hbar^2}m^*|J\Omega|^{2/3}(\Delta\varepsilon)^{1/3},$$

which represent the energy, the magnetic field, the length of the radius vector of the electron, the radius of the polaron state, the reciprocal concentration of magnetic ions, and the band g-factor, respectively. The case $J<0$ is also described by functional (8), once we replace σ by $-\sigma$ and $\varkappa$ by $-\varkappa$. That case therefore does not require a separate discussion. With $h=0$, expression (8) becomes

$$\varepsilon_\sigma(\rho)=\frac{1}{\rho^2}-\frac{1}{u}\rho^3\int_0^\infty\left\{\left[1+\frac{1}{\rho^6}\exp(-4y^2)\right]^{1/2}-1\right\}y^2\,dy. \tag{10}$$

It can be seen in particular from expression (10) that the polaron energy ε_σ, thought of as a function of the radius of the state, ρ, is positive both as $\rho\to 0$ and as $\rho\to\infty$. In other words, a self-localization is not favored from the energy standpoint for such values of ρ. At finite values of ρ, the quantity ε_σ may go negative if the dimensionless concentration of the magnetic ions, $1/u$, is sufficiently high. The self-localized states are separated from the band states by an energy barrier,[24] and the equation determining the extrema, $d\varepsilon_\sigma(\rho)/d\rho=0$, may have two roots in addition to the trivial solution $\rho=\infty$, which corresponds to a delocalized state of an electron. These two other roots describe a minimum and a local maximum of the energy near the barrier. In the case $h\neq 0$, solutions of this sort can appear for each of the spin states of the polaron, for which the equation for determining the minimum becomes

$$u=\frac{6}{\rho}\int_0^\infty dy\left\{\left[\frac{1}{4}-h^2+\frac{1}{4}(1+4h^2)^{1/2}\left(1+4\left(h-\frac{\sigma}{\rho^3}\exp(-2y^2)\right)^2\right)^{1/2}+\frac{h\sigma}{\rho^3}\exp(-2y^2)\right]y^2\exp(-4y^2)\Big/\left[1+4\left(h-\frac{\sigma}{\rho^3}\exp(-2y^2)\right)^2\right]^{1/2}\left[(1+4h^2)^{1/2}+\left(1+4\left(h-\frac{\sigma}{\rho^3}\exp(-2y^2)\right)^2\right)^{1/2}\right]^2\right\}. \tag{11}$$

With $h=0$ we find

$$u=\frac{3}{2\rho}\int_0^\infty\frac{y^2\exp(-4y^2)\,dy}{[1+\exp(-4y^2)/\rho^6]^{1/2}\{1+[1+\exp(-4y^2)/\rho^6]^{1/2}\}}. \tag{12}$$

Figure 1 shows $\rho(u)$ curves according to Eq. (11) for various values of h. It follows from this figure that there is a

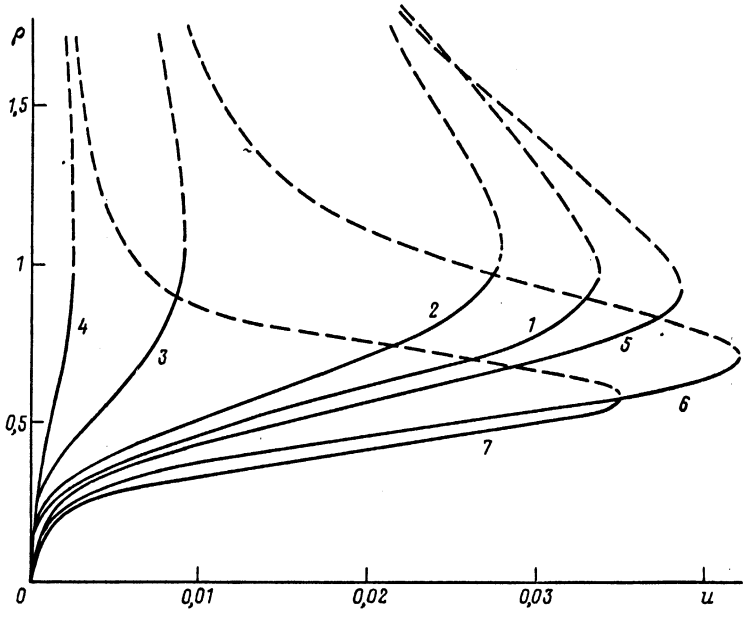

FIG. 1. Radius of the polaron state, ρ, versus the reciprocal concentration of the magnetic component, u, according to Eq. (11). 2–4—$\sigma=-1/2$, $h=0.1$ and 0.5; 5–7—$\sigma=1/2$, with the same values of h; 1—$h=0$. The parts of the curves shown by dashed lines describe states which correspond to the crests of the self-localization barrier.

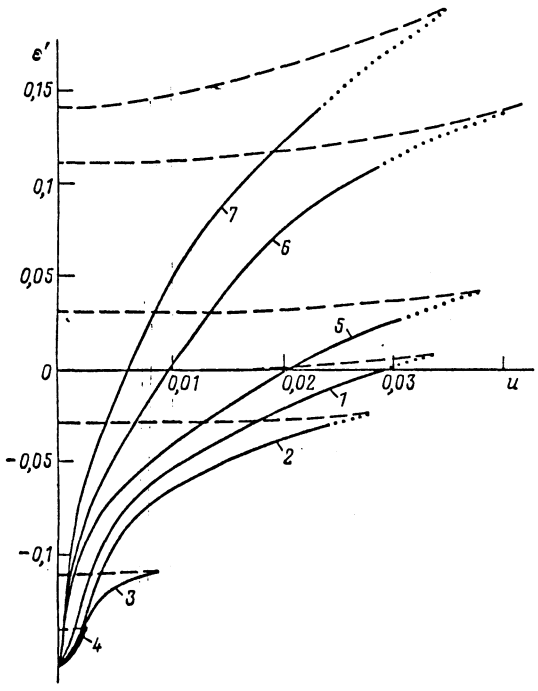

FIG. 2. The energies $\varepsilon' = (\varepsilon - \varkappa h\sigma)u$ in the region of a stable state (solid lines) and in that of an unstable state (dotted lines) of a polaron and near the maximum of the self-localization barrier (dashed line) versus the reciprocal concentration of the magnetic impurity, u, for the values of h and σ in Fig. 1. By virtue of the particular choice of origin for the scales of $\varkappa$, h, and σ, and by virtue of the normalization to a concentration $1/u \propto n_I$, the self-localization energy is reckoned from the value $\varepsilon'_\infty = (\varepsilon_\infty - \varkappa h\sigma)u$, to which the energy of the barrier tends in the limit $u \to 0$, according to (13).

certain critical impurity concentration $n_{cr} \propto 1/u_{cr}$ such that a polaron can exist only at $n > n_{cr}$ ($u < u_{cr}$). This quantity depends on the magnetic field and the spin projection. Analysis shows that the branches of the $\rho(u)$ curves for $\rho < \rho_{cr} = \rho(u_{cr})$ correspond to a local minimum of energy (8), while the branches of these curves at $\rho > \rho_{cr}$ correspond to a local maximum of ε_σ, i.e., to an energy barrier separating the band states of the electron from the polaron states.

To determine which values $u < u_{cr}$ and $\rho < \rho_{cr}$ correspond to absolute minima of functional (8), i.e., to stable states of a polaron, we use Eq. (11) to plot $\varepsilon_\sigma = \varepsilon_\sigma(u)$ (Fig. 2). The solid lines in Fig. 2 show the depth of the local minimum of $\varepsilon = \varepsilon(\rho)$; the dashed lines show the height of the energy barrier. With increasing concentration ($u \to 0$), this height decreases rapidly, and it reaches the energy of a delocalized electron:

$$\varepsilon_\infty = \varkappa h\sigma + \left[\frac{4h}{(1+4h^2)^{1/2}}\right]\frac{\sigma c}{u}, \qquad (13)$$

where $c = (\pi/128)^{1/2}$. If we go over to dimensional variables in (13), using (9), we find the standard expression in the theory of the spin splittings of a Van Vleck semimagnetic semiconductor:[26]

$$E_\infty = g_e\mu_B H\sigma - Jx\langle S_{Iz}\rangle\sigma, \qquad (14)$$

where $\langle S_{Iz}\rangle$ is the spin angular momentum of one magnetic ion in the field **H**, which is determined by the expression in square brackets in (13).

It can be seen from a comparison of ε_σ and ε_∞ that for values of h close to zero, and for $\sigma = 1/2$, the critical concentration ($1/u^{**}$) for the appearance of an energy barrier and a local minimum is quite different from the concentration ($1/u^*$) which corresponds to a stable state of a polaron. For $\sigma = -1/2$ and $h > 1$, the quantities u^* and u^{**} are practically the same. We also note that the stability region of states with $\sigma = 1/2$ is wider than that for $\sigma = -1/2$, despite the fact that the magnetization is directed opposite the magnetization of the crystal as a whole in the electron self-localization region. With increasing h, the difference becomes larger. This circumstance is reflected on the "phase diagrams" (Fig. 3), which show the regions of values of h and u for which self-localized states exist and the regions in which such states exist and are furthermore stable (the hatched regions in Fig. 3).

The curves of $\varepsilon(u)$ and $\rho(u)$ found here are, along with the boundaries of the stability region of the self-localized states (Fig. 3), the basic characteristics of the polaron state. They determine several quantities which can be observed experimentally. In the following sections of this paper, we calculate some of these quantities.

4. THE *g*-FACTOR OF A POLARON

In research dating back to the pioneering studies of Refs. 27 and 28, it has been established that at low temperatures the energy of spin transitions for band electrons in a semimagnetic semiconductor is determined primarily by the carrier-ion exchange interaction. This interaction is de-

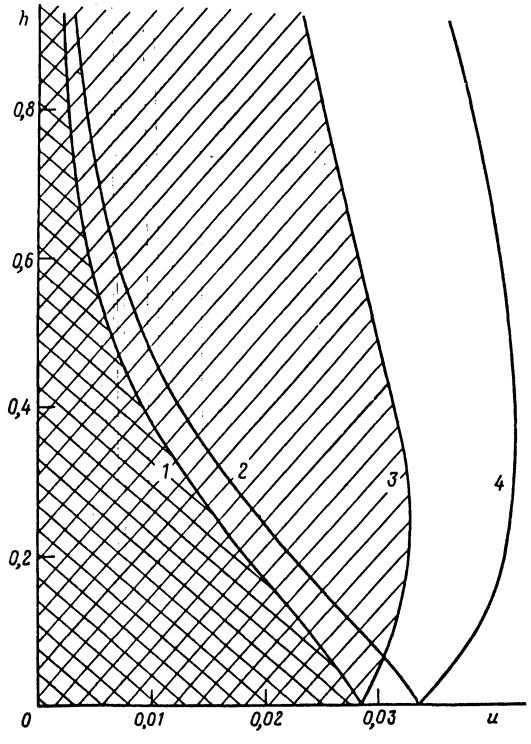

FIG. 3. Diagram of polaron states. The region bound by curves *1* and *2* corresponds to unstable states for $\sigma = -1/2$; the region bounded by curves *3* and *4* corresponds to unstable states for $\sigma = 1/2$. The hatched regions to the left of curves *1* and *3* correspond to stable states for the values $\sigma = -1/2$ and $\sigma = 1/2$, respectively.

scribed in the mean-field approximation by the second term in (14). The effective g-factor corresponding to this splitting which is found from the relation

$$\hbar\omega_s = E_{1/2} - E_{-1/2} = g_{eff}\mu_B H, \tag{15}$$

is proportional to the concentration of magnetic ions (if they dominate the situation). This proportionality remains in force for localized large-radius electron states. When the polaron effect is taken into account, however, the n_I dependence of the g-factor is more complex.[10] One might expect that in this case we would again find a deviation of $g_{\text{eff}}(n_I)$ from the behavior imposed by (14): $g_{\text{ex}} = 4g_I Jx/\Delta\varepsilon$. Evidently a measure of this deviation is the quantity $\eta = (g_{\text{eff}} - g_e)/g_{\text{ex}}$, where, according to (15), we have

$$g_{eff} = \frac{1}{\mu_B}\left[\frac{\partial(\hbar\omega_s)}{\partial H}\right]_{H=0}. \tag{16}$$

Substituting (8) into (15), and then into (16), we find, in terms of the dimensionless variables in (9),

$$\eta = \frac{16\rho^3}{(2\pi)^{1/2}} \times \int_0^\infty \frac{\exp(-2y^2)\{2\rho^3[\rho^3+(\rho^6+\exp(-4y^2))^{1/2}]+\exp(-4y^2)\}y^2\,dy}{[\rho^6+\exp(-4y^2)]^{1/2}\{\rho^3+[\rho^6+\exp(-4y^2)]^{1/2}\}^2}. \tag{17}$$

Figure 4 shows a plot of $\eta = \eta(u)$ according to (17) for that branch of the $u(\rho)$ curve which corresponds to stable polarons. We find a characteristic feature of nonlinear systems with saturation: $g_{\text{eff}} - g_e < g_{\text{ex}}$. The reason for this feature is that the exchange field induced by the field $\mathbf{H}$ in accordance with (14) and the self-action exchange field due to the polaron effect do not make additive contributions to the electron energy. We also see that this effect strengthens as the polaron effect increases with increasing n_I (with decreasing u).

5. MAGNETIC MOMENT OF A POLARON

The self-localization of an electron is accompanied by the appearance of a correlation between the spins of magnetic ions, induced by an electron spin $\mathbf{S}_e$ involved in an exchange interaction with them. As a result, a spin angular momentum $\mathbf{S}_\Sigma$ and a related magnetic moment $\mathbf{M} = -g_I\mu_B\mathbf{S}_\Sigma$ are associated with a polaron. The appearance of a free magnetic polaron is thus accompanied by an increase in the magnetization of the crystal in proportion to the number of polarons.

By definition we have

$$S_{\Sigma z} = n_I \int \langle 1|S_{Iz}|1\rangle dV, \tag{18}$$

where $|1\rangle$ is the eigenvector corresponding to the lowest-lying level of Hamiltonian (2), and the matrix element itself is, according to the theorem regarding the derivative of an energy with respect to a parameter,[29] $dE_1(g_I\mu_B H + A\sigma)/d(g_I\mu_B H)$, where E_1 is given in (5). Since we are interested not in the magnetization itself but in its change $\Delta\mathbf{M}(\propto \Delta S_\Sigma)$ associated with the formation of a polaron, we should subtract from (18) the corresponding quantity corresponding to the case $J = 0$. Carrying out some calculations, and going over to dimensionless variables in (9), we find

$$\mathcal{M} = \frac{\sigma\rho^3}{u}\int_0^\infty \left[\frac{h-\sigma\exp(-2y^2)/\rho^3}{\{1+4[h-\sigma\exp(-2y^2)/\rho^3]^2\}^{1/2}} - \frac{h}{(1+4h^2)^{1/2}}\right] y^2\,dy, \tag{19}$$

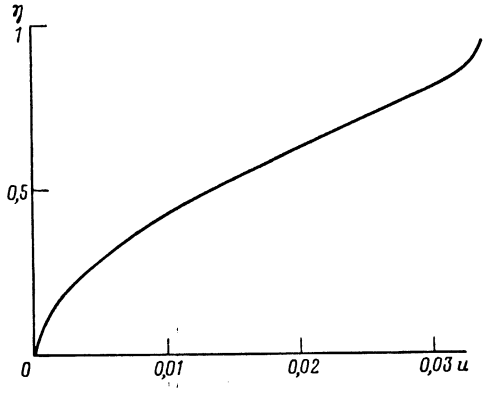

FIG. 4. The factor $\eta = (g_{\text{eff}} - g_e)/g_{\text{ex}}$, which determines the relative weakening of the exchange contribution to the effective polaron g-factor in comparison with the exchange contribution of a free electron (or hole). These results are shown for weak magnetic fields [Eq. (17)]. The independent variable is the reciprocal concentration of the magnetic component.

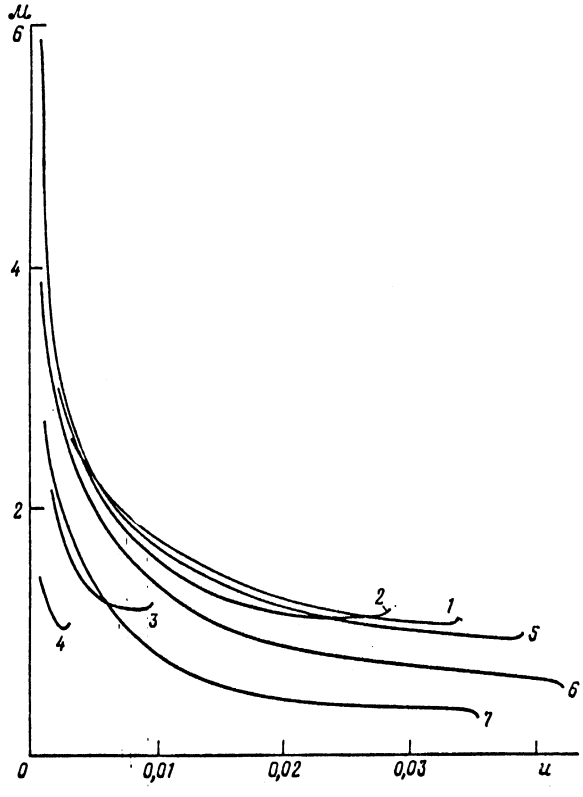

FIG. 5. Magnetic moment of a polaron according to expressions (19) and (20) versus the reciprocal concentration of the magnetic component. Curves *1*, *2*,..., correspond to the values of h and σ specified in the Fig. 1 caption.

This quantity is related to the spin of a polaron by

$$\mathcal{M}=\Delta S_z m^* |J\Omega|^{3/2} (\Delta\varepsilon)^{1/2}/3\pi^2\hbar^2. \quad (20)$$

Figure 5 shows the result of a numerical evaluation of expression (19). We see that the magnetic moment of a polaron increases slightly with increasing concentration of magnetic ions, despite the decreasing radius of the state (Fig. 1). We also note that at $u \approx u^*$ the spin of a polaron, S_Σ, for $\sigma = 1/2$ is considerably smaller than that for $\sigma = -1/2$. The reason for this difference is that in the case $\sigma = 1/2$, in which the local magnetization in the self-localization region is directed opposite the magnetization of the overall crystal in the field **H**, the potential-well depth required for self-localization can be reached more quickly than in the case $\sigma = -1/2$, in which these magnetizations are in the same direction. This circumstance is also responsible for the lower critical concentrations $1/u^{**}$ for polarons with $\sigma = 1/2$.

6. EFFECTIVE MASS OF A POLARON

Many important characteristics of a semiconductor are related to the effective mass of the polaron, m^{**} (Ref. 22). In particular, at sufficiently large effective masses m^{**} the polarons localize at potential fluctuations and do not take part in charge transport.

In calculating m^{**} we make use of an idea described in Ref. 30. According to that idea, as a polaron undergoes a translational motion it perturbs the medium in which it has formed. It perturbs the medium as it would if its wave vector were $\psi_t = \psi_{e,v}(x - vt, y, z)$ (where the x axis runs along the direction of motion). As a result of this perturbation, the energy of the polaron acquires an increment v^2. The coefficient of this increment is by definition $m^{**}/2$. It is a considerably more complex matter to implement that idea here than in Ref. 30, however, because the nonlinearity of the susceptibility of the Van Vleck semimagnetic semiconductor, which we are taking into account, shapes the basic characteristics of the polaron. To go beyond the scope of the linear-response approximation in calculating m^{**}, we use the tools of adiabatic perturbation theory.[31] It is assumed in that theory that a perturbation which has an adiabatically slow time dependence sends the system from its ground state (which corresponds to a polaron at rest) into excited states, with a probability amplitude proportional to the time derivative of the perturbation. As a result, the wave function of ion j can be written as a linear combination of the functions of the ground state $|1\rangle_t$ and the excited states $|2\rangle_t, \ldots$, calculated as stationary states for a fixed value of the variable perturbation (not necessarily a small value) at some given time t:

$$|G\rangle = \alpha|1\rangle_t + \beta|2\rangle_t + \ldots, \quad |\alpha|^2 + |\beta|^2 + \ldots = 1. \quad (21)$$

The average energy of ion j corresponding to state (21) is

$$E_G^j = \langle G|\mathcal{H}_l^j|G\rangle = |\alpha|^2 E_1^j(t) + |\beta|^2 E_2^j(t) + \ldots,$$

where $E_i^j(t)$ is the energy of the ith stationary state of the ion found in the adiabatic limit. Using normalization (21), we find

$$E_G^j = E_1^j(t) + \hbar\omega_{21}|\beta|^2 + \ldots,$$

where $\hbar\omega_{i1} = E_i - E_1$. The quantity $|\beta|^2$ is the probability (P_{12}) for a transition from the state $|1\rangle$ to the state $|2\rangle$ and is given by[31]

$$P_{1i} = \left| \int_{-\infty}^{t} \left(\frac{\partial \mathcal{H}_l^j(t_1)}{\partial t_1} \right) \frac{1}{\hbar\omega_{i1}(t_1)} \exp\left[i \int_{-\infty}^{t_1} \omega_{i1}(\tau) d\tau \right] dt_1 \right|^2. \quad (22)$$

The increment in the energy due to the motion of the polaron is therefore

$$\Delta E = E_G^j - E_1^j(t) = \sum_i \hbar\omega_{i1} P_{1i}.$$

Summing over the magnetic ions in the continuum approximation, we find the following expression for the effective mass:

$$m^{**} = \frac{2n_l}{v^2} \sum_i \int \hbar\omega_{i1} P_{1i} d^3\mathbf{r}. \quad (23)$$

To find an explicit expression for P_{1i}, we go over to the variable $\xi = x - vt$ in the integrals in (22), and we use

$$(\mathcal{H}_l^j)_{mn} = \int (S_{lz}^j)_{mn} dA, \quad (24)$$

where A was introduced in Eq. (6). In this case we find from (22)

$$P_{1i} = \left| \int_{\xi}^{\infty} (S_{lz})_{i1} \frac{\partial A}{\partial x} \frac{1}{\hbar\omega_{i1}} \exp\left(\frac{i}{v} \int_{x}^{\infty} \omega_{i1}(x') dx' \right) dx \right|^2. \quad (25)$$

It can be shown (see Appendix 1) that we have

$$\lim_{v\to 0} \frac{1}{v^2} \left| \int_{\xi}^{\infty} f(x) \exp\left[\frac{i}{v} \varphi(x) \right] dx \right|^2 = \left| \frac{f(\xi)}{\varphi'(\xi)} \right|^2, \quad (26)$$

where $\varphi'(\xi) = d\varphi/d\xi$. For P_{1i} we then find

$$P_{1i}|_{v\to 0} \to v^2 \left| (S_{lz})_{i1} \frac{\partial A}{\partial x} \frac{1}{\hbar\omega_{i1}} \right|^2_{x=\xi} \Big/ |\omega_{i1}(\xi)|^2. \quad (27)$$

From this point on the calculations depend on the particular model adopted for the center, which is determined by Hamiltonian (2). Also making use of our assumption 5 above, we can show that the only nonvanishing matrix element of the operator S_{lz} in (24) is $(S_{lz})_{21}$, where $|2\rangle_t$ corresponds to the first excited state of the magnetic ion. Using expression (5) and the results of Appendix 2, we find

$$\hbar\omega_{21} = [(\Delta\varepsilon)^2 + 4(g_l\mu_B H + A\sigma)^2]^{1/2}, \quad (S_{lz})_{21} = -\frac{2^{1/2}\Delta\varepsilon}{\hbar\omega_{21}}. \quad (28)$$

The calculation of the effective mass now reduces to an expansion of the energy of this system, $E_G^j = E_1^j + \Delta E$, in a series in the velocity, in which we retain terms up to $\sim v^2$. For this purpose, we substitute an expansion of the wave function $\psi_{e,v} = \psi_e + v^2\psi_1$ into the expression for the energy. It can be shown (Ref. 30; see also Appendix 3) that the contribution of the correction ψ_1 to ΔE is of order higher than v^2, so the effective mass is given by expressions (23), (27), and (28), in which we substitute the wave function of a polaron at rest, (7). Going through this procedure, we find

$$m^{**}=\frac{16\pi}{3}n_l(\Delta\varepsilon)^2\hbar^2\int_0^\infty\left(\frac{\partial A}{\partial r}\right)^2\frac{r^2\,dr}{[(\Delta\varepsilon)^2+4(g_l\mu_B H+A\sigma)^2]^{5/2}},\tag{29}$$

In deriving (29) we used $\partial A/\partial x=(\partial A/\partial r)(\partial r/\partial x)$, and we integrated over angles. Introducing the dimensionless variables in (9), we finally find

$$\mu=\frac{1}{u\rho^5}\int_\infty^0\frac{y^4\exp(-4y^2)\,dy}{\{1+4[h-\sigma\exp(-2y^2)/\rho^3]^2\}^{5/2}},\tag{30}$$

where

$$\mu=m^{**}m^*|J\Omega|^{4/3}(\Delta\varepsilon)^{7/3}/8\pi^2\hbar^4,$$

and ρ is related to u by the equation which determines the extremum of $\varepsilon_\sigma(u)$, (10), whose solution is shown in Fig. 1. Figure 6 shows calculated curves of $\mu(u)$. We see that the effective mass of the polaron increases monotonically with increasing concentration of magnetic ions, for all values of h and σ. With increasing magnetic field, the polaron mass corresponding to the lowest-lying energy branch, with $\sigma=-1/2$, falls off sharply, while for $\sigma=1/2$ the value of m^{**} increases significantly at a given value of the concentration $1/u$. The reason for this result is the small difference between the magnetizations inside and outside the self-localization region, as we mentioned above. The perturbation of the medium by the moving polaron is thus smaller in the case $\sigma=-1/2$ than in the case $\sigma=1/2$. With $h=0.5$ and $u=0.007$, for example, the effective masses of the two polaron branches differ by about two orders of magnitude.

7. DISCUSSION OF RESULTS; CONCLUSION

We have shown that a free magnetic polaron can in principle exist in a Van Vleck semimagnetic semiconductor.

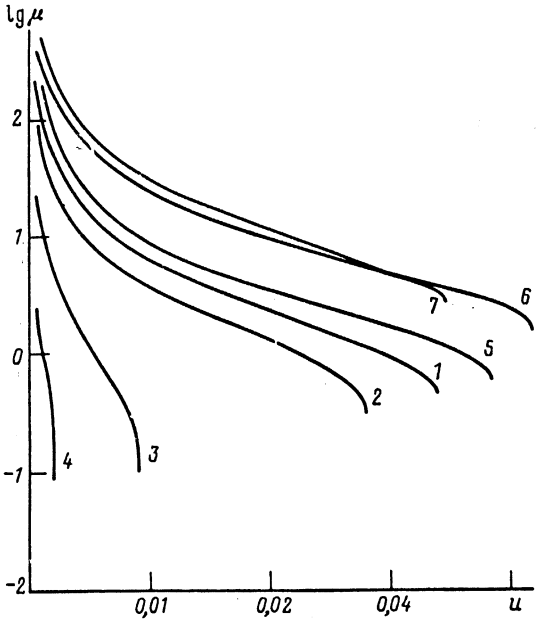

FIG. 6. Effective polaron masses determined from expression (30) versus the reciprocal concentration of the magnetic component. Curves *1*, *2*,... correspond to the values of h and σ used in Fig. 1.

Let us look at some numerical estimates of the critical concentration, using as an example the solid solution $Zn_{1-x}Fe_xSe$, for which we have $J_e=0.22$ eV, $J_h=-1.6$ eV, $\Omega=45.6\cdot10^{-24}$ cm^3, $\Delta\varepsilon=1.8$ meV, $m_e^*=0.14m_0$, and $m_h^*=1.2m_0$ (Refs. 14 and 32), where m_0 is the mass of a free electron. An estimate of $x_{cr}=n_{cr}\Omega$ from the value $u_{cr}(H=0)=0.0337$ yields $x_{cr}\approx8.6>1$ for a band electron. In other words, the electron cannot undergo self-localization. For a hole, in contrast, we would have $x_{cr}\approx0.037$, so we could expect manifestations of free magnetic polarons in effects involving holes.

We should stipulate that we are ignoring the complex nature of the valence band and the particular features of the splitting of this band in the effective exchange field. The latter factor has the consequence, for example, that a spherically symmetric single-parameter function as in (7) is not a good approximation of $\psi(\mathbf{r})$. Analysis of a corresponding problem for an orientational semimagnetic semiconductor of the $A^2_{1-x}Mn_xB^6$ type has shown[33] that incorporating the complex structure of the Γ_8 valence band does not lead to order-of-magnitude changes in the basic characteristics of a polaron. On this basis we suggest that the estimates presented here convey the basic features of a free magnetic polaron of the hole type in a Van Vleck semimagnetic semiconductor in a qualitatively correct way. Refining these estimates is an independent problem, which goes beyond the scope of the present paper.

Estimating the radius of the state on the basis of the calculated curves in Fig. 1 and the relationship between ρ and a in (9), we easily find $a=2.7\cdot10^{-7}$ cm with $u=u_{cr}$ and $H=0$ for the parameter values listed above. This estimate shows that a polaron forms in a large-radius state in the sense $\overline{N}=\frac{4}{3}\pi a^3(n_l)_{cr}\gg1$ in this crystal. This result justifies our assumption 2. We also see that the latter inequality is not violated at any value of σ, as h and $n_l(1/u)$ increase.

We turn now to possible experimental manifestations of the polarons discussed above and ways to excite such polarons. The first place to seek a contribution from a free magnetic polaron is in time-varying effects. The polaron might be manifested by a shift of the luminescence line of free excitons, which exist as Wannier excitons in Van Vleck semimagnetic semiconductors. Over its lifetime, a hole bound in an exciton (or the overall exciton, by virtue of the hole) may tunnel through the barrier which separates the polaron states from the band states, form a resonant state, and then relax to a (self-localized) polaron state[24] (as was shown above, an electron cannot undergo self-localization). A calculation of the probability for a tunneling of this sort goes beyond the scope of the present paper. However, this probability is finite, and it contributes to the luminescence at a frequency shifted from the free-exciton band by an amount equal to the energy of the polaron.

This scenario for the formation of a polaron may prevail for holes in the lowest-lying energy state, with $\sigma=-1/2$. For holes with $\sigma=1/2$, in contrast, either a polaron can form directly in this excited state, or there should first be a spin flip and then a relaxation to a polaron state. The presence of polarons in Van Vleck semimagnetic semiconductors is thus manifested as an energy shift of the emitted light during the luminescence of a free exciton. The shift is toward longer wavelengths.

The presence of a free magnetic polaron may lead to the appearance of a new band in the exciton reflection spectra of Van Vleck semimagnetic semiconductors as the result of a direct optical excitation of polaron states.

A free magnetic polaron can also be observed in experiments on electron spin resonance, in which the free magnetic polaron would be manifested as a corresponding shift of the g-factor (as discussed above). Furthermore, free magnetic polarons should exhibit superparamagnetism effects associated with the large value of the polaron spin, S_Σ, in measurements of the magnetic susceptibility or magnetization of the Van Vleck semimagnetic semiconductor. A calculation from (20) with the data in Fig. 5 shows that the relation $S_\Sigma \geqslant 16$ holds in this crystal. In other words, these effects should be manifested even in very weak magnetic fields.

Another experimental manifestation of a free magnetic polaron might be a contribution (or, more precisely, the essentially total absence of a contribution) to charge transport in Van Vleck semimagnetic semiconductors. In strong magnetic fields, the charge transport in a Van Vleck semimagnetic semiconductor is by band carriers. When polarons arise in weak fields H under the condition $n_l > n_{cr}$, with a mobility far lower because of the larger effective mass, the electrical conductivity of such semiconductors should decrease. For the effective mass of a polaron of the hole type in $Zn_{1-x}Fe_xSe$, estimates yield

$$m^{**}=8{,}4\cdot 10^3\, m_0\mu, \tag{31}$$

where μ is given by (29) or by the curves in Fig. 6. Using $\mu = 0.1$ as an estimate, we find that a free magnetic polaron is heavier by a factor of 840 than a free electron and is therefore practically excluded from transport phenomena. The contribution of free magnetic polarons to transport phenomena in Van Vleck semimagnetic semiconductors thus reduces to a jump in the electrical conductivity when the boundaries of the region of (h, u) values shown on the "phase diagram" in Fig. 3 are crossed.

Our analysis has been carried out for $T = 0$. This simplification is justified by the weak temperature dependence of the magnetic susceptibility of Van Vleck semimagnetic semiconductors at $T \ll \Delta\varepsilon$ (Ref. 15). As the temperature is raised, however, the susceptibility decreases; this decrease will result in a decrease in the probability for the formation of new polaron states and in the destruction of existing polaron states.

What would happen if we discarded the assumptions which we made in deriving the energy of a free magnetic polaron in (4)? The spin-spin interaction between magnetic impurities may lead to certain quantitative changes. In particular, in A^2FeB^6 Van Vleck semimagnetic semiconductors, this interaction will reduce the magnetic susceptibility of the system. This effect requires large critical concentrations for the formation of free magnetic polarons; i.e., it reduces the probability for the formation of such entities. This effect can be taken into account in a semiquantitative way by renormalizing $\Delta\varepsilon$ to the value of Θ ($\Theta > 0$ for an antiferromagnetic exchange interaction), which plays a role analogous to the Curie-Weiss parameter in ordinary orientation magnetic materials. Composition fluctuations, whose relative magnitude in a sphere with a radius equal to the radius of the polaron state in the Gaussian case, $\propto \overline{N}^{-1/2}$, lead to fluctuations in the energy of the polaron. The latter fluctuations will affect all the experimental manifestations discussed above, without exception. In particular, because of the large effective mass, these fluctuations will prevent the motion of free magnetic polarons and will completely suppress the polaron contribution in transport phenomena.

In conclusion we would like to point out that in a magnetic field there could be other polaron states, in addition to the free magnetic polarons which we discussed here. These other states would be realized if the magnetic length $a_H = (\hbar c/eH)^{1/2}$ were smaller than the polaron radius. They would be quasi-one-dimensional because of the special direction (that of the magnetic field). They would not be separated from the band states by an energy barrier. Rewriting the condition $a > a_H$ in terms of ρ and h, and using data for $Zn_{1-x}Fe_xSe$, we find

$$\rho(u, h) \geqslant 2{,}38/h^{1/5}, \tag{32}$$

In other words, even in weak magnetic fields there is a large region of ρ values in which polarons of this type exist. A study of these polarons is of interest for both theory and experiment.

In summary, it has been shown here that the possibility of a self-localization of free charge carriers, primarily holes, should be taken into consideration in studies of magnetic, optomagnetic, and transport phenomena in Van Vleck semimagnetic semiconductors with high concentrations of magnetic impurities. Manifestations of this self-localization might be the features discussed above in the magnetic-field, concentration, and temperature dependence of the processes under study.

APPENDIX 1

We consider

$$\alpha = \int_\xi^\infty f(x)\exp\left[\frac{i}{v}\varphi(x)\right]dx. \tag{A1}$$

Introducing $x - \xi = t$ in (A1), we find

$$\alpha = \int_0^\infty f(t+\xi)\exp\left[\frac{i}{v}\varphi(t+\xi)\right]dt. \tag{A2}$$

Introducing $t = vz$ in (A2), we find

$$\alpha = v\int_0^\infty f(vz+\xi)\exp\left[\frac{i}{v}\varphi(vz+\xi)\right]dz, \tag{A3}$$

In the low-velocity limit we then find

$$\alpha = v\int_0^\infty f(\xi)\exp\left[\frac{i}{v}(\varphi(\xi)+vz\varphi'(\xi))\right]dz$$

$$= vf(\xi)\exp\left[\frac{i}{v}\varphi(\xi)\right]\int_0^\infty \exp[iz\varphi'(\xi)]dz, \tag{A4}$$

where $\varphi'(\xi) = d\varphi/d\xi$. The integral $\int_0^\infty \exp[iz\varphi'(\xi)]dz$ is evaluated by multiplying the integrand by $\exp(\eta z)$ $\int_0^\infty (\eta > 0)$ and using the identity[34]

$$\frac{1}{\eta - i\varphi'(\xi)}\Big|_{\eta\to 0} = \frac{i}{\varphi'(\xi)} + \pi\delta(\varphi'(\xi)). \tag{A5}$$

If $\varphi'(\xi)$ does not vanish anywhere on the real axis (and in our case we have $\varphi' = \omega_{21} > 0$), the second term in (A5) is zero, and we can write

$$|\alpha|^2 = v^2 |f(\xi)|^2 / |\varphi'(\xi)|^2. \tag{A6}$$

We have thus proved Eq. (26) of the text proper.

APPENDIX 2

For a T_d symmetry, the matrix of Hamiltonian (2), which incorporates the ground state for the case $\mathbf{H} \| [001]$ in the basis $|A_1\rangle$, $|T_1\rangle$, $|E\rangle$, given in Ref. 35, is

$$\hat{\mathcal{H}} = -2^{1/2} g_l \mu_B H \hat{S}_z + \begin{pmatrix} 0 & 0 & 0 \\ 0 & \Delta\varepsilon & 0 \\ 0 & 0 & 2\Delta\varepsilon \end{pmatrix}, \quad \hat{S}_z = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}. \tag{A7}$$

A diagonalization leads to a ground-state energy E_1 as given by (5), while the energies of the excited states are found to be

$$E_2 = \Delta\varepsilon, \quad E_3 = \Delta\varepsilon + [(\Delta\varepsilon)^2 + 4(g_l \mu_B H)^2]^{1/2}, \tag{A8}$$

This diagonalization also leads to the corresponding eigenvectors $|u_1\rangle$, $|u_2\rangle$, and $|u_3\rangle$. Taking the matrix elements of the operator $\hat{S}_z$ between $|u_1\rangle$ and $|u_2\rangle$ and also between $|u_1\rangle$ and $|u_3\rangle$, we find $(S_z)_{13} = 0$, while $(S_z)_{12}$ is given by Eq. (28) of the text proper.

APPENDIX 3

We consider an energy functional of a moving polaron:

$$E = \int \left\{ \frac{\hbar^2}{2m^*} (\nabla\psi)^2 + W[\psi] + W_1[\psi] \right\} dx\, dy\, dz, \tag{A9}$$

where the first two terms determine a polaron at rest. We substitute ψ in the form

$$\psi = \psi_0 + v^2 \psi_1 + \dots \tag{A10}$$

into this expression, and we expand E, retaining terms up to those on the order of v^2. We find

$$E = \int \left\{ \frac{\hbar^2}{2m^*} (\nabla\psi_0)^2 + v^2 \frac{\hbar^2}{m^*} \nabla\psi_0 \nabla\psi_1 + W[\psi_0] + v^2 \psi_1 \frac{\partial W}{\partial \psi_0} + v^2 W_1[\psi_0] \right\} dx\, dy\, dz. \tag{A11}$$

The terms which do not depend on v determine the energy of a polaron at rest and are of no interest. Let us examine an expansion of the energy of a polaron at rest in which we retain terms up to $\sim v^2$:

$$E_1 = v^2 \int \left\{ \frac{\hbar}{m^*} \nabla\psi_0 \nabla\psi_1 + \psi_1 \frac{\partial W}{\partial \psi_0} \right\} dx\, dy\, dz. \tag{A12}$$

Integrating the first term in (A12) by parts, we find

$$I = \int \left[\frac{\partial\psi_0}{\partial x}\frac{\partial\psi_1}{\partial x} + \frac{\partial\psi_0}{\partial y}\frac{\partial\psi_1}{\partial y} + \frac{\partial\psi_0}{\partial z}\frac{\partial\psi_1}{\partial z} \right] dx\, dy\, dz$$

$$= -\int \psi_1 \left[\frac{\partial^2\psi_0}{\partial x^2} + \frac{\partial^2\psi_0}{\partial y^2} + \frac{\partial^2\psi_0}{\partial z^2} \right] dx\, dy\, dz. \tag{A13}$$

Substituting (A13) into (A12), we find that the expression in braces in (A12) is an equation which determines the structure of a polaron at rest:

$$E_1 = v^2 \int \psi_1 \left[\frac{\hbar^2}{m^*} \Delta\psi_0 + \frac{\partial W}{\partial \psi_0} \right] dx\, dy\, dz. \tag{A14}$$

The latter expression vanishes identically when we substitute the function ψ_0 into it. This vanishing proves the assertion that ψ_1 does not contribute to m^{**}.

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