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Theory of photo-magnetization of an interacting particle system: application to $Hg_{1-x}Mn_xTe$

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Abstract

We derive a theory of light-induced magnetization of an interacting particle system, which describes a model diluted magnetic semiconductor. We set up a many-body Hamiltonian that includes electron and hole kinetic energies, carrier–photon and carrier–local-moment interactions and all the Coulomb interactions. Using Heisenberg equations of motion and a mean-field approximation, we derive expressions for the carrier spin density and excitonic amplitudes. Non-equilibrium relaxation is included phenomenologically. The interdependence of carrier spin density and the local-moment magnetization is calculated self-consistently. We apply our theory of photo-magnetization to $Hg_{1-x}Mn_xTe$, choosing parameters of the model appropriate to this system. Our results for the photo-magnetization versus the input laser power agree qualitatively with the observed trends in this system. We also study the photo-magnetization as a function of temperature, manganese concentration and photon energy and the results obtained are along expected lines.

1. Introduction

The interplay between light and magnetism through magneto-optic effects such as the Faraday rotation and the Kerr effect has been well known for a long time [1]. The former refers to the rotation of the plane of polarization per unit distance of the linearly polarized light propagating along the direction of the magnetic field in a crystal; the latter, on the other hand, describes the conversion of plane polarized light into an elliptical polarization on reflection along the magnetization direction, which is a high symmetry direction, by the surface of a magnetic crystal. However, photo-magnetization or light-induced magnetization has currently emerged as an interesting and important area in condensed matter research. Indeed it has been observed in a variety of systems such as cyanometalate-based magnets [2–6], spin crossover complexes [7], diluted magnetic semiconductors [8, 9] and their quantum wells [10]. Other materials of interest in this regard are doped manganites [11], spinel ferrite films [12] and organic-based magnets [13]. Of late, ferromagnetic semiconductors have attracted a great deal of attention [14].

In the present work we focus on the photo-induced paramagnetization in diluted magnetic semiconductors, particularly $Hg_{1-x}Mn_xTe$. It would be pertinent, therefore, to review the experimental situation for this case. Krenn *et al* [8] reported the observation of light-induced magnetization in $Hg_{1-x}Mn_xTe$ in the paramagnetic limits by a technique combining optical pumping and a superconducting interference detector. They showed that the magnetization appears due to the orientation of Mn ions caused by spin transfer from polarized electrons and holes to Mn centers in the process of spin-flip exchange scattering. A detailed report of the experiments by the same group followed later [9], who analyzed the optically induced magnetization both in narrow gap $Hg_{1-x}Mn_xTe$ and wide gap $Cd_{1-x}Mn_xTe$ [15] subjected to circularly polarized light. Their analyses of Mn-spin orientations by carriers include two mechanisms: (i) the static polarization induced by the mean field of spin-polarized electrons and (ii) the dynamic polarization caused by the s–d

spin-flip scattering. The latter was shown to be suppressed by Mn–Mn interactions. They also presented several systematics concerning the light-induced magnetization in $Hg_{1-x}Mn_xTe$ such as photo-magnetization versus laser power for fixed photon energy, photo-magnetization versus exciting photon energy and photo-magnetization versus temperature for fixed laser power and frequency. They ascribed that the photo-magnetization occurs in the absence of an external magnetic field in the following way. Circularly polarized light creates spin-polarized carriers, which in turn align the Mn ions, thus creating magnetization. Efforts are made by the authors of the experimental work to explain their own observations by semi-empirical methods. However, their mechanisms lack the rigor of many-body analyses of the phenomena.

Recently, we considered this problem [16], starting from a many-body Hamiltonian for a system of photo-generated electrons and holes, which are spin-split by magnetic ions in the diluted magnetic semiconductor. An expression for the photo-magnetization was obtained, following an equation of motion method, as a function of the photon power and the frequency. Damping of non-equilibrium carriers and spin excitons was considered in a phenomenological way. The drawbacks of the model are as follow. All the many-body interactions such as electron–electron, hole–hole and electron–hole interactions are not considered adequately. No attempt has been made to develop a self-consistent magnetic field from the interaction of the light field with the semiconductor and the interactions between the carriers and the local magnetic moments. Further, the theory was not parameterized in order to reproduce some experimentally observed quantity.

Motivated by the above remarks and in view of the current interest in the carrier-induced magnetization in the magnetic and diluted magnetic semiconductors with regard to their importance in spintronics, we decided to extend our previous works further. In section 2, starting from a many-body Hamiltonian for the electrons and holes, carrier–light field, carrier–local-moment and all the Coulomb interactions, we derive nonlinear equations for the carrier spin density. The interdependence of the carrier spin density and the local-moment magnetization is evaluated self-consistently. Finally, we parameterize our model within reasonable physical limits to quantify our theoretical calculation so as to compare with a part of the experimental observations in the case of narrow gap $Hg_{1-x}Mn_xTe$ [9]. This is the concluding section of our work, which also discusses our results *vis-à-vis* some of the experimental findings

2. Theory of light-induced magnetization

2.1. Model Hamiltonian

We consider the following Hamiltonian for the diluted magnetic semiconductor subject to a light field as

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\rm int} + \mathcal{H}_{\rm c} + \mathcal{H}_{ls}, \tag{1}$$

where different terms are explained as follows:

$$\mathcal{H}_0 = \sum_{\vec{k}\sigma} (\varepsilon^{\rm e}_{\vec{k}} + E_{\rm g}) c^{\dagger}_{\vec{k}\sigma} c_{\vec{k}\sigma} - \sum_{\vec{k}\sigma} \varepsilon^{\rm h}_{-\vec{k}} d^{\dagger}_{-\vec{k}\sigma} d_{-\vec{k}\sigma}, \tag{2}$$

In the above, $\mathcal{H}_0$ describes the Hamiltonian for band electrons and holes. $\varepsilon^{\rm e}_k$ and $\varepsilon^{\rm h}_k$ are the energies of an electron and a hole, respectively; $E_{\rm g}$ is the bandgap; ($c^{\dagger}$ and c) and ($d^{\dagger}$ and d) are the creation and annihilation operators for electrons and holes, respectively. σ represents the spin index and takes both up- and down-spin states for the carriers. σ and $-\sigma$ are opposite spin states, i.e. for $\sigma = \uparrow$, $-\sigma = \downarrow$ and vice versa.

The second term in equation (1), which describes the important exchange interaction between the electron and the local moments, is obtained from the equation

$$\mathcal{H}_{\rm int} = \tfrac{1}{2} \sum_j J(\vec{r}) \vec{\sigma} \cdot \vec{S}_j, \tag{3}$$

where $(1/2)\vec{\sigma}$ is the electron spin operator and $\vec{S}_j$ is the spin of the magnetic impurity at the jth site. J is the exchange interaction and $\vec{\sigma}$ are the Pauli spin matrices. Ignoring the spin fluctuation terms, the interaction Hamiltonian is written as

$$\mathcal{H}_{\rm int} = \tfrac{1}{2} x N J(r) \sigma_z S_z, \tag{4}$$

where the subscript z denotes the z components of the respective operators. In equation (4), a virtual crystal approximation, which is a standard practice in semiconductors, is considered. Here x is the fraction of magnetic ions and N is the total number of cations. Assuming an average exchange interaction, we obtain the many-body Hamiltonian for the electron-local-moment interaction as

$$\mathcal{H}_{\rm int} = \tfrac{1}{2} x \langle S_z \rangle \sum_{n={\rm c,v}} J_n \sum_{k\eta,\eta'} c^{\dagger}_{n\vec{k}\eta} \sigma_{z\eta\eta'} c_{n\vec{k}\eta'} \tag{5}$$

where c and v stand for conduction and valence bands, respectively. Here both η and η' take $\uparrow$ and $\downarrow$ spin states. Simplification of equation (5) leads to

$$\mathcal{H}_{\rm int} = \tfrac{1}{2} x \langle S_z \rangle \sum_k [J_{\rm c}(c^{\dagger}_{{\rm c}k\uparrow} c_{{\rm c}k\uparrow} - c^{\dagger}_{{\rm c}k\downarrow} c_{{\rm c}k\downarrow}) + J_{\rm v}(c^{\dagger}_{{\rm v}k\uparrow} c_{{\rm v}k\uparrow} - c^{\dagger}_{{\rm v}k\uparrow} c_{{\rm v}k\uparrow})]. \tag{6}$$

Assuming that the annihilation of a valence band electron at $\vec{k}$ is equivalent to the creation of a hole with $-\vec{k}$, and vice versa, i.e. $c_{{\rm v}\vec{k}} = d^{\dagger}_{-\vec{k}}$ and $c^{\dagger}_{{\rm v}\vec{k}} = d_{-\vec{k}}$ we write the above equation in terms of electron and hole operators as

$$\mathcal{H}_{\rm int} = \tfrac{1}{2} x \langle S_z \rangle \sum_{\vec{k}} \{J_{\rm e}(c^{\dagger}_{\vec{k}\uparrow} c_{\vec{k}\uparrow} - c^{\dagger}_{\vec{k}\downarrow} c_{\vec{k}\downarrow}) - J_{\rm h}(d^{\dagger}_{-\vec{k}\uparrow} d_{-\vec{k}\uparrow} - d^{\dagger}_{-\vec{k}\downarrow} d_{-\vec{k}\downarrow})\}, \tag{7}$$

where $J_{\rm c}$ was replaced by $J_{\rm e}$. We have neglected the antiferromagnetic local-moment couplings.

The third term in equation (1) is given by

$$\mathcal{H}_{\rm c} = \tfrac{1}{2} \sum_{\vec{q}\vec{k}\vec{k}'\sigma\sigma'} V(\vec{q}) c^{\dagger}_{\vec{k}+\vec{q}\sigma} c^{\dagger}_{\vec{k}'-\vec{q}\sigma'} c_{\vec{k}'\sigma'} c_{\vec{k}\sigma} + \tfrac{1}{2} \sum_{\vec{q}\vec{k}\vec{k}'\sigma\sigma'} V(\vec{q}) d^{\dagger}_{\vec{k}+\vec{q}\sigma} d^{\dagger}_{\vec{k}'-\vec{q}\sigma'} d_{\vec{k}'\sigma'} d_{\vec{k}\sigma} - \tfrac{1}{2} \sum_{\vec{q}\vec{k}\vec{k}'\sigma\sigma'} V(\vec{q}) c^{\dagger}_{\vec{k}+\vec{q}\sigma} d^{\dagger}_{\vec{k}'-\vec{q}\sigma'} d_{\vec{k}'\sigma'} c_{\vec{k}\sigma}. \tag{8}$$

$\mathcal{H}_{\rm c}$ describes the Coulomb (electron–electron (e–e), hole–hole (h–h) and electron–hole (e–h)) interactions. $V(q)$ is the Fourier transform of the Coulomb energy.

The fourth term in equation (1) is

$$\mathcal{H}_{ls} = G\sum_{\vec{k}\alpha}(c^{\dagger}_{k\alpha}d^{\dagger}_{-\vec{k}-\alpha}B + d_{-\vec{k}-\alpha}c_{\vec{k}\alpha}B^{\dagger}). \tag{9}$$

Equation (9) describes the other important interaction of the formulation, which is the interaction of the light field with the semiconductor. $B^{\dagger}$ and B are photon creation and annihilation operators. G is an averaged coupling strength between the semiconductor and the light field; $\alpha = \pm$ denoting $\alpha \pm$ circular polarization of light, which fixes the spin configurations of the photo-excited electrons and holes [17]. For simplicity, we assume α to be $1/2$ both for electrons and holes. Thus these are treated on a par with σs and are replaced by them in subsequent occurrences We consider only resonant terms, i.e. an electron–hole pair is created by the absorption of a photon or an electron–hole pair recombines with the emission of a photon. In other words, energy non-conserving terms are neglected. This is known as the rotating wave approximation. Both excitonic ($c^{\dagger}d^{\dagger}, dc$) and photon operators are time-dependent. It may be noted that photo-induced ferromagnetism has recently been studied without any specific polarization of the classical light field [18, 19].

We consider a mean-field approximation (MFA) and write in the first order

$$H = \sum_{\vec{k}\sigma}\varepsilon^{\mathrm{e}}_{\vec{k}\sigma}c^{\dagger}_{\vec{k}\sigma}c_{\vec{k}\sigma} - \sum_{\vec{k}\sigma}\varepsilon^{\mathrm{h}}_{-\vec{k}\sigma}d^{\dagger}_{-\vec{k}\sigma}d_{-\vec{k}\sigma} + G\sum_{\vec{k}\sigma}(c^{\dagger}_{\vec{k}\sigma}d^{\dagger}_{-\vec{k}-\sigma}B + d_{-\vec{k}-\sigma}c_{\vec{k}\sigma}B^{\dagger}), \tag{10}$$

where

$$\varepsilon^{\mathrm{e}}_{\vec{k}\sigma} \cong \varepsilon^{\mathrm{e}}_{\vec{k}} + E_{\mathrm{g}} \pm \frac{1}{2}xJ_{\mathrm{e}}\langle S_z\rangle + \frac{1}{N}\langle\{[c_{\vec{k}\sigma}, \mathcal{H}_{\mathrm{c}}], c^{\dagger}_{\vec{k}\sigma}\}\rangle \tag{11}$$

and

$$\varepsilon^{\mathrm{h}}_{\vec{k}\sigma} \cong \varepsilon^{\mathrm{h}}_{\vec{k}} \pm \frac{1}{2}xJ_{\mathrm{h}}\langle S_z\rangle + \frac{1}{N}\langle\{[d_{\vec{k}\sigma}, \mathcal{H}_{\mathrm{c}}], d^{\dagger}_{\vec{k}\sigma}\}\rangle. \tag{12}$$

The positive sign in the third term of equation (11) and the second term in equation (12) and elsewhere in this paper is taken for $\sigma = \uparrow$ and the negative sign for $\sigma = \downarrow$. In equations (11) and (12)

$$\langle\{[c_{\vec{k}\sigma}, \mathcal{H}_{\mathrm{c}}], c^{\dagger}_{\vec{k}\sigma}\}\rangle = -\sum_{\vec{q}}V(\vec{q})n^{\mathrm{e}}_{\vec{k}-\vec{q}\sigma} + V(0)\sum_{\vec{k}}(n^{\mathrm{e}}_{\vec{k}\sigma} + n^{\mathrm{e}}_{\vec{k}-\sigma}) - V(0)\sum_{\vec{k}}(n^{\mathrm{h}}_{\vec{k}\sigma} + n^{\mathrm{h}}_{\vec{k}-\sigma}) \tag{13}$$

and

$$\langle\{[d_{\vec{k}\sigma}, \mathcal{H}_{\mathrm{c}}], d^{\dagger}_{\vec{k}\sigma}\}\rangle = -\sum_{\vec{q}}V(\vec{q})n^{\mathrm{h}}_{\vec{k}-\vec{q}\sigma} + V(0)\sum_{\vec{k}}(n^{\mathrm{h}}_{\vec{k}\sigma} + n^{\mathrm{h}}_{\vec{k}-\sigma}) - V(0)\sum_{\vec{k}}(n^{\mathrm{e}}_{\vec{k}\sigma} + n^{\mathrm{e}}_{\vec{k}-\sigma}). \tag{14}$$

If we assume that the carriers are only intrinsic and Mn^{2+} spins contribute no carriers, the last two terms in both equations (15) and (16) cancel because the number of photo-generated electrons would be equal to the corresponding number of holes, and we have, from equations (11)–(14),

$$\varepsilon^{\mathrm{e}}_{\vec{k}\sigma} \cong \varepsilon^{\mathrm{e}}_{\vec{k}} + E_{\mathrm{g}} \pm \frac{1}{2}xJ_{\mathrm{e}}\langle S_z\rangle - \frac{1}{N}\sum_{\vec{q}}V(\vec{q})n^{\mathrm{e}}_{\vec{k}-\vec{q}\sigma} \tag{15}$$

and

$$\varepsilon^{\mathrm{h}}_{\vec{k}\sigma} \cong \varepsilon^{\mathrm{h}}_{\vec{k}} + E_{\mathrm{g}} \pm \frac{1}{2}xJ_{\mathrm{h}}\langle S_z\rangle - \frac{1}{N}\sum_{\vec{q}}V(\vec{q})n^{\mathrm{h}}_{\vec{k}-\vec{q}\sigma} \tag{16}$$

where $n^{\mathrm{e}}_{\vec{k}\sigma} = \langle c^{\dagger}_{\vec{k}\sigma}c_{\vec{k}\sigma}\rangle$ and $n^{\mathrm{h}}_{\vec{k}\sigma} = \langle d^{\dagger}_{\vec{k}\sigma}d_{\vec{k}\sigma}\rangle$. It may be noted, in passing, that Mn^{2+} ions act as acceptors in some (III, Mn)V semiconductors, thus contributing to both itinerant and local-moment magnetism [14].

2.2. Equations of motion

In order to derive equations of motion for the carrier density and excitonic amplitudes, we consider Heisenberg's equation of motion of the type

$$\mathrm{i}\hbar\frac{\mathrm{d}Q}{\mathrm{d}t} = [Q, H], \tag{17}$$

where Q is a time-dependent operator. From equations (10) and (17), we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}(\hat{n}^{\mathrm{e}}_{\vec{k}\sigma} + \hat{n}^{\mathrm{h}}_{\vec{k}-\sigma}) = -\frac{2\mathrm{i}}{\hbar}G(\hat{A}^{\dagger}_{\vec{k}\sigma-\sigma}B - \hat{A}_{\vec{k},\sigma-\sigma}B^{\dagger}) \tag{18}$$

and

$$\frac{\mathrm{d}}{\mathrm{d}t}\hat{A}^{\dagger}_{\vec{k}\sigma-\sigma} = \frac{\mathrm{i}}{\hbar}[(\varepsilon^{\mathrm{e}}_{\vec{k}\sigma} - \varepsilon^{\mathrm{h}}_{\vec{k}-\sigma})\hat{A}^{\dagger}_{\vec{k}\sigma-\sigma} - G(\hat{n}^{\mathrm{e}}_{\vec{k}\sigma} + \hat{n}^{\mathrm{h}}_{-\vec{k}-\sigma} - 1)B^{\dagger}]. \tag{19}$$

Further $\hat{n}^{\mathrm{e}}_{\vec{k}\sigma} = c^{\dagger}_{\vec{k}\sigma}c_{\vec{k}\sigma}$, $\hat{n}^{\mathrm{h}}_{\vec{k}\sigma} = d^{\dagger}_{\vec{k}\sigma}d_{\vec{k}\sigma}$ and $\hat{A}^{\dagger}_{\vec{k}\sigma-\sigma} = c^{\dagger}_{\vec{k}\sigma}d^{\dagger}_{-\vec{k}-\sigma}$, where the hat sign above n and A denotes the operator nature of the quantities.

Since we are interested in the variation of the magnetization as a function of the light field which is assumed to be a classical one, B and $B^{\dagger}$ are replaced respectively by their average values β and β^{*}. We also replace the number density and excitonic operators by their average values defined as classical variables. We neglect quantum fluctuations and assume a mean-field decoupling of the type $\langle PQ\rangle = \langle P\rangle\langle Q\rangle$.

We set

$$\sum_{\vec{q}}n^{\mathrm{e,h}}_{\vec{k}-\vec{q}\sigma} = \sum_{\vec{k}}n^{\mathrm{e,h}}_{\vec{k}\sigma}, \tag{20}$$

which holds good for a spatially homogeneous system by the conservation of momentum [20]. Further we assume an average inter-particle interaction and replace $(1/N)\sum_{\vec{q}}V(\vec{q})$ by U. The damping of the carrier and excitonic number densities is considered in a phenomenological way. With the above approximations, equations (17) and (19) can be written as

$$\frac{\mathrm{d}}{\mathrm{d}t}(n^{\mathrm{e}}_{\vec{k}\sigma} + n^{\mathrm{h}}_{-\vec{k}-\sigma}) = -\frac{2\mathrm{i}}{\hbar}G(A^{\dagger}_{\vec{k}\sigma-\sigma}\beta - A_{\vec{k},\sigma-\sigma}\beta^{*}) + \gamma^{\mathrm{c}}_{\sigma-\sigma}(n^{\mathrm{e}}_{\vec{k}\sigma} + n^{\mathrm{h}}_{-\vec{k}-\sigma}) \tag{21}$$

and

$$\frac{\mathrm{d}}{\mathrm{d}t}A^{\dagger}_{\vec{k}\sigma-\sigma} = \frac{\mathrm{i}}{\hbar}[\{E_{\mathrm{g}} + xJ\langle S_z\rangle - U(n^{\mathrm{e}}_{-\vec{k}\sigma} - n^{\mathrm{h}}_{-\vec{k}-\sigma})\}A^{\dagger}_{\vec{k}\sigma-\sigma} - G(n^{\mathrm{e}}_{\vec{k}\sigma} + n^{\mathrm{h}}_{-\vec{k}-\sigma} - 1)\beta^{*}] + \gamma^{\mathrm{eh}}_{\vec{k}\sigma-\sigma}A^{\dagger}_{\vec{k}\sigma-\sigma}. \tag{22}$$

In the above equations the number density and excitonic operators are replaced by their mean values (without hats).

$J = J_e + J_h$. γ^c and γ^{eh} are the damping of the carrier number and spin exciton amplitude, respectively. The damping terms result from incoherent processes. Since the damping mechanism is statistical in nature and is due to random processes, it is included phenomenologically. While the phenomenological treatment is certainly not valid under all circumstances it has nevertheless been found to represent in many cases the proper qualitative or even quantitative descriptions of the observed phenomena. This phenomenological procedure can be justified from first principles by coupling the system to a heat bath. Further, in equation (22), there would be a change of sign in the $xJ\langle S_z\rangle$ term upon spin exchange (i.e. when $\sigma = \downarrow$ and $-\sigma = \uparrow$).

We now introduce explicit time dependence of the excitonic and photon field variables and assuming that they have the same frequency dependence, ω, we write $A_{\vec{k}\sigma-\sigma} = \tilde{A}_{\vec{k}\sigma-\sigma}e^{-i\omega t}$ and $\beta_{\vec{k}\sigma-\sigma} = \tilde{\beta}_{\vec{k}\sigma-\sigma}e^{-i\omega t}$. Substituting these in equations (21) and (22), and considering only stationary solutions, we obtain

$$\tilde{A}^{\dagger}_{\vec{k}\sigma-\sigma} = \frac{G(n^e_{\vec{k}\sigma} + n^h_{-\vec{k}-\sigma} - 1)\tilde{\beta}^*}{\{E_g - \hbar\omega + xJ\langle S_z\rangle - U(n^e_{-\vec{k}\sigma} - n^h_{-\vec{k}-\sigma})\} - i\hbar\gamma^{eh}_{\vec{k}\sigma-\sigma}} \tag{23}$$

and

$$\gamma^c_{\sigma-\sigma}(n^e_{\vec{k}\sigma} + n^h_{-\vec{k}-\sigma}) = \frac{2i}{\hbar}G(\tilde{A}^{\dagger}_{\vec{k}\sigma-\sigma}\tilde{\beta} - \tilde{A}_{\vec{k},\sigma-\sigma}\tilde{\beta}^*). \tag{24}$$

We assume

$$n^e_{\vec{k}\sigma} + n^h_{-\vec{k}-\sigma} \cong n^e_{-\vec{k}\sigma} - n^h_{-\vec{k}-\sigma} \cong f^e_{\vec{k}\sigma} \tag{25}$$

i.e. these are replaced by their average values $f^e_{\vec{k}\sigma}$. Thus, with the help of equations (23) and (25), we obtain from equation (24)

$$f^e_{\vec{k}\sigma} = \frac{4(\gamma^{eh}_{\sigma-\sigma}/\gamma^c_{\sigma-\sigma})G^2(1 - f^e_{\vec{k}\sigma})|\tilde{\beta}|^2}{[E_g - \hbar\omega + xJ\langle S_z\rangle - Uf^e_{\vec{k}\sigma}]^2 + (\gamma^{eh}_{\sigma-\sigma})^2}. \tag{26}$$

Denoting $\sum_{\vec{k}} f^e_{\vec{k}\sigma} = n_\sigma$ and assuming that the damping parameters remain constant under spin exchange, i.e. $\gamma^c_{\uparrow\downarrow} = \gamma^c_{\downarrow\uparrow} = \gamma^c$ and $\gamma^{eh}_{\uparrow\downarrow} = \gamma^{eh}_{\downarrow\uparrow} = \gamma^{eh}$, which is valid in a mean-field approximation, the up-and down-electron spin densities can be written as

$$n_{\uparrow(\downarrow)} = \frac{a(1 - n_{\uparrow(\downarrow)})n_{ph}}{1 + \{b(\omega, x) + cn_{\uparrow(\downarrow)} \pm xd\}^2}, \tag{27}$$

where $n_{ph} = |\tilde{\beta}|^2$ and is proportional to the photon density and the dimensionless parameters are as follows: $a = 4G^2/\hbar^2\gamma^{eh}\gamma^c$, $b(\omega, x) = (E_g - \hbar\omega)/\hbar\gamma^{eh} = b_2(x) - b_1(\omega)$, $b_2(x) = E_g(x)/\hbar\gamma^{eh}, b_1(\omega) = \hbar\omega/\hbar\gamma^{eh}, c = -U/\hbar\gamma^{eh}$ and $d = J\langle S_z\rangle/\hbar\gamma^{eh}$. It may be noted that there is interdependence between the local-moment magnetization, which is proportional to $\langle S_z\rangle$, and the carrier spin density, $m_s(n_\uparrow - n_\downarrow)$. These are related by

$$\langle S_z\rangle = (1 - x)^{12}SB_s(\xi), \tag{28}$$

where $B_s(\xi)$ is the Brillouin function:

$$B_s(\xi) = \frac{2S+1}{2S}\coth\left(\frac{2S+1}{2S}\xi\right) + \frac{1}{2S}\coth\left(\frac{\xi}{2S}\right), \tag{29}$$

and the other quantities are $\xi = yS$, $y = g\mu_B\mathcal{H}_o/k_BT$ and $\mathcal{H}_o = m_sJ/2g\mu_B$. In equation (28), $(1-x)^{12}$ is the probability of finding a single magnetic ion with only non-magnetic ions as its neighbors in an fcc lattice and $S = \frac{5}{2}$ for Mn^{2+} ions. The system in question, $Hg_{1-x}Mn_xTe$, crystallizes in the zinc-blende structure. Since the conventional cell is fcc, where for a given cation there are 12 nearest cation neighbors, this number appears in the power in equation (28).

Equation (28) is a nonlinear equation for the up-and down-spin electron densities. The nonlinearity is due to the occurrence of $n_{\uparrow(\downarrow)}$ in the second term as well as in the last term of the squared quantity in the denominator. In the absence of the Coulomb term, there is still nonlinearity and finite spin density. However, the spin density is zero if $d = 0$, implying that the carrier–local-moment interaction is essential for the spin density. Further, if $a = 0$, implying that the light coupling is zero, both $n_\uparrow$ and $n_\downarrow$ are zero separately, which is due to the fact that the spin-polarized carriers are generated by the photons only. Thus the photo-magnetization of an itinerant system can be independent of Coulomb interactions. A finite Coulomb interaction can, however, be used to fine-tune the results. Further temperature dependence is considered only through the Brillouin function. It can also be incorporated via the Fermi distribution function $f^e_{\vec{k}\sigma}$, but at the temperatures of interest, its effect would be small.

Equation (27) is solved self-consistently to obtain the spin density $(n_\uparrow - n_\downarrow)$. Analytic expressions for the spin density, m_s, can be obtained if we neglect the $(cn_{\uparrow(\downarrow)})^2$ term in the denominator of equation (27) in the small c approximation. In that case the equation for m_s is quadratic. This approximation is discussed in our previous work [16]. The solutions of equation (27) are obtained in the paramagnetic limit.

The light–carrier–local-moment interaction creates an effective magnetic field, which acts both on the carriers and the local moments. In the presence of such an interaction, the photo-magnetization is given by [9]

$$M = -\tfrac{1}{2}\mu_B g_{eff}(n_\uparrow - n_\downarrow). \tag{30}$$

where g_{eff} is the effective g factor in the presence of the spin–orbit interaction and carrier–local-moment interaction [21, 22]. The photon power is calculated from

$$P_L = n_{ph}\int_0^{\omega_m}\hbar\omega_{ph}\,d\omega_{ph}, \tag{31}$$

where ω_m is the maximum photon frequency considered.

3. Results and discussion

Our results are plotted in figures 1–4. We calculate the photo-magnetization as a function of photon power, using equations (27), (30) and (31). The parameters considered are: $G = 1$ meV, $\gamma^c = 5.16\times10^6\ s^{-1}$, $\gamma^{eh} = 1.03\times10^{13}\ s^{-1}$, $E_g = 0.213$ eV, $U = 0.019$ eV and $J = -0.026$ eV. The values

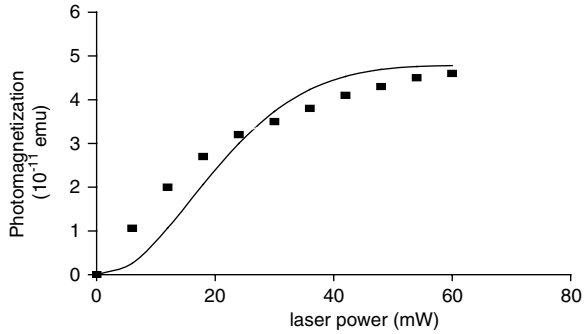

Figure 1. Photo-magnetization (10^{-11} emu) versus laser power (mW). Parameters used are: $G = 1$ meV, $\gamma^{c} = 5.16 \times 10^{6}$ s^{-1}, $\gamma^{eh} = 1.03 \times 10^{13}$ s^{-1}, $E_g = 0.213$ eV, $U = 0.019$ eV, $J = -0.026$ eV, $x = 0.135$, $\hbar\omega = 232$ meV, $p = p_{av}$. Experimental points (■) are from [9].

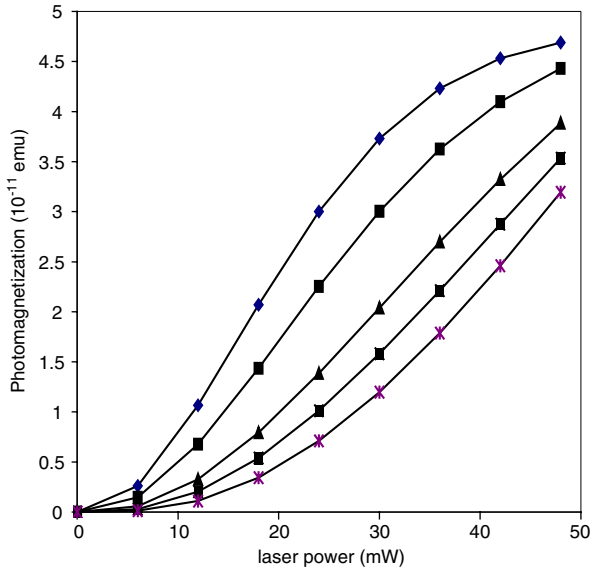

Figure 2. Photo-magnetization (10^{-11} emu) versus laser power (mW) for different temperatures. The parameters used are as in figure 1. Photo-magnetization decreases with increase in temperature for a fixed laser power. The curves in decreasing order are for temperatures 4.2, 5.2, 7.5, 10 and 15 K, respectively.

(This figure is in colour only in the electronic version)

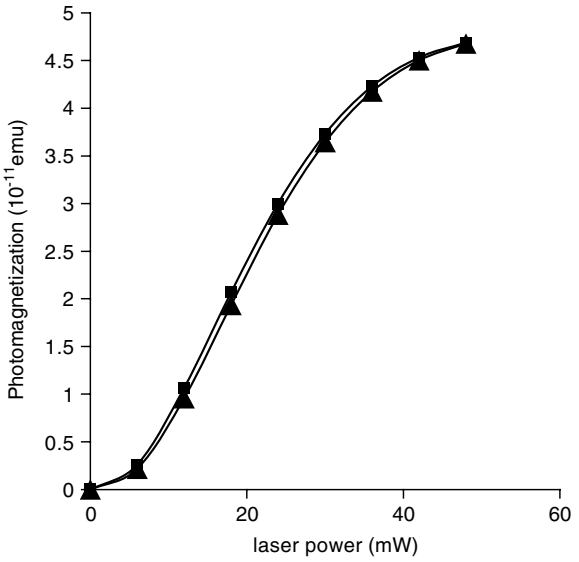

Figure 3. Photo-magnetization (10^{-11} emu) versus laser power (mW) for two typical values of x (fraction of manganese ions) $= 0.132$ and 0.135 (in increasing order). With increase in x, photo-magnetization increases. Other parameters are as in figure 1.

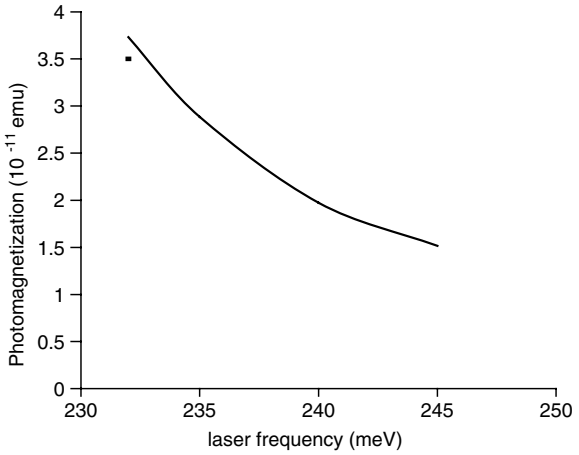

Figure 4. Photo-magnetization (10^{-11} emu) versus laser frequency (meV). Laser power chosen is 30 mW. Other parameters are as in figure 1. The shaded box denotes the experimental value for $\hbar\omega = 232$ meV. The decrease in photo-magnetization is inconsistent with experimental observation [9].

of manganese concentration $x(0.135)$ and the energy gap, E_g, are taken from values considered by the experiments [9]. The value of G, the light semiconductor coupling energy, is of the order of the attainable Rabi splitting. The relaxation parameters γ^{c} and γ^{eh} correspond to relaxation times 2×10^{-7} s and 10^{-13} s, respectively. Thus damping of excitons is larger than the damping of the spin-polarized carriers. In other words, the lifetime of excitons is a few orders of magnitude smaller than the lifetime of the spin-polarized carriers. This should be the case, otherwise there would have been no magnetization, if the spin-polarized carriers had damped faster than the excitons. Indeed the damping ratio of the spin excitons to the spin-polarized carriers is of the order of 10^{7}, implying that the damping of the carriers is almost negligible compared to the exciton damping. The excitonic lifetime is of the order of picoseconds ($\approx 10^{-13}$ s). The experimental relaxation times are also of this order [9].

The value of the Coulomb energy and the exchange energy are expected to be reasonable for the system. Within the paramagnetic phase of the diluted magnetic semiconductor, particularly for the system under study, the magnitude of U appears to be justified. Exact experimental values for these are not available. Photon energy is considered greater than the bandgap. The effective g value g_{eff} is taken equal to 100, because small energy gap and large spin–orbit interaction in $Hg_{1-x}Mn_xTe$ can give large g factors. This is also the value considered by the experimenters. Thus, parameters chosen tally with the values available from the experiment [9]

Table 1. Photo-magnetization (10^{-11} emu) versus laser power (mW) for different approximations. In the second column the calculated results for the photo-magnetization are presented when the probability (p) of finding the magnetic ion is p_1 i.e. $p_1 = (1-x)^{12}$. The corresponding results in the third and fourth columns are for $p = p_2 = 1$ and $p = p_{\rm av} = \frac{1}{2}(p_1 + p_2)$, respectively. The experimental results are in the fifth column.

	Photo-magnetization (10^{-11} emu)			
Laser power (mW)	p_1	p_2	$p_{\rm av}$	$p_{\rm exp}$ [9]
0	0	0	0	0
6	0.05	1.448	0.261	1.06
12	0.386	2.932	1.066	2.0
18	0.987	4.006	2.069	2.7
24	1.698	4.739	3.0	3.2
30	2.397	5.105	3.731	3.5
36	3.024	5.188	4.231	3.8
42	3.568	5.13	4.531	4.1
48	4.035	5.018	4.69	4.3
54	4.435	4.894	4.759	4.5
60	4.778	4.778	4.778	4.6

in $Hg_{1-x}Mn_xTe$. We have compared our results with the corresponding values of the experiment [9] in figure 1. We find reasonably fair agreement between our theory and the experiment. The subject assumes added importance in view of the fact that a fervent search is going on to find carrier-induced ferromagnetism in semiconductors, which would be of importance in the futuristic spintronic and computer industry.

We shall now discuss our results. In equation (28), we used the probability of finding magnetic ions in the fcc lattice. However, with this value of probability, our results show a decrease of magnetization *vis-à-vis* the experimental values. The decrease is quite considerable up to the laser power of 30 mW. On the other hand, if we take the probability as unity, our values for the magnetization are higher than the experimental values. Therefore, we have considered an effective probability which is the average of these two probabilities and the results are plotted in figure 1, using this average probability, which shows reasonably good agreement with experimental values. This is done, of course, at the cost of self-consistency. The accuracy of the probability function depends on the nature of the disorder due to the magnetic impurities. Thus the treatment requires improvement over the virtual crystal approximation. These results are presented in table 1 in detail. The self-consistency is considered up to five decimal places.

In figure 2 we have considered the temperature variations of the photo-magnetization as a function of laser power. With the increase in temperature the photo-magnetization decreases, as it should. The temperature dependence is taken into account via the Brillouin function. It may be noted that the experimentalists have not shown any transition to a ferromagnetic state. Hence the temperature variation is consistent with that observed in a paramagnetic system. As mentioned earlier, the maximum power considered by the experimenters corresponds to a magnetic field of the order of about a hundredth of an Oersted, which is much less than the typical exchange field responsible for ferromagnetism. It may be of interest to note, in passing, that light-induced magnetization contributes to a very small increase in T_c in a model (III, Mn)V system [19].

In figure 3, we have plotted the photo-magnetization versus laser power for two magnetic-ion concentrations. With the decrease in x, the photo-magnetization decreases, for a given strength of laser power. This can be understood as follows. With the decrease in x, the local-moment concentration decreases and hence a reduction in the local-moment magnetization. On the other hand, with the decrease in x, the bandgap decreases, and hence the carrier density should increase for a given laser power, increasing thereby the magnetization. In the competition between these two mechanisms, the former has an edge and thus we see a decrease in the magnetization.

In figure 4, we have plotted the photo-magnetization as a function of laser frequency for a fixed laser power 30 mW. The observed trend is consistent with experiment for $\hbar\omega \geqslant 232$ meV because our parameters, chosen to plot figure 1, are obtained for a photon energy of 232 meV. This may also presumably be due to an increase in the exciton condensate with an increase in the photon frequency, thereby decreasing the spin density. This is also apparent from equation (26). With the increase in $\hbar\omega$, there is an increase in the magnitude of b, thus causing a decrease in $n_\uparrow$ and $n_\downarrow$.

In summary, we set up a many-body Hamiltonian, which includes carrier kinetic energies, carrier–local-moment and carrier–light-field interactions and all the Coulomb interactions. We evaluate Heisenberg's equations of motion for the carrier densities and excitonic amplitudes in a mean-field approximation. Non-equilibrium relaxations are considered phenomenologically. The equations are solved and nonlinear equations are obtained for $n_\uparrow$ and $n_\downarrow$ as functions of photon density. The interdependence of the carrier spin density and the local magnetic moment proportional to $\langle S_z \rangle$ is calculated self-consistently. Temperature dependence is included through the Brillouin function. Our results, based on a reasonable set of parameters, agree with experiment on qualitative grounds. The application of the theory to the system under study is mostly qualitative and does not take into account the detailed physics of the system.

There is still scope for considerable improvement of the present work. Instead of considering the effective g value in an ad hoc way, we can calculate it through a $\vec{k}\cdot\vec{\pi}$ ($\vec{\pi}$ being the momentum operator in the presence of spin–orbit interaction) model that could be developed on a set of pre-calculated energy levels for HgMnTe or HgTe. If this is done, the two-band approximation can be improved and electron and hole dispersions can be incorporated. Although, we considered the photon frequency greater than the bandgap to suit the narrow gap HgMnTe there is, we believe, nothing in this theory which restricts it only to this case. The model can also be explored for wide bandgap semiconductors with energy gaps larger than the photon frequency. With regard to the phenomenological use of the damping parameters, we wish to make the following comments. If the system is in contact with a heat bath, then after sufficient time has elapsed it must reach a state which is, generally speaking, not stationary, since the external field depends on time, but which is independent

of the initial conditions. This inconsistency can be avoided by following the Keldysh approach [23]. These would be for future work and we shall report on it when the results are ready.

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