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On the nonequilibrium light-induced diamagnetism

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Abstract. A new interpretation of light-induced magnetization changes of a magnetic semiconductor, manganese arsenide (MnAs), observed by the authors of references [1,2], is proposed in this paper. Contrary to references [1,2], where the results of experiments were interpreted as the observation of light-induced phase transition, here we propose a completely different approach. It suggests that at least far from the vicinity of T_c, there are no real magnetization changes as in case of phase transition, but there are changes of the magnetic flux threading the MnAs-sample. These changes are due to non-equilibrium light-induced diamagnetic moments of quasi-free electrons of narrow d-subbands of the MnAs-conduction band. The other aspects of the experiments of [1,2] are also discussed and some similarity between this effect and the orbital diamgnetism due to persistent currents in mesoscopic structures is emphasised.

PACS. 75.50.Pp Magnetic semiconductors – 75.20.Ck Nonmetals

1 Introduction

In the papers [1,2] the observation of an interesting phenomenon, light-induced magnetization change of the magnetic semiconductor manganese arsenide, was reported. As is known [3], under normal conditions manganese arsenide (MnAs) is a ferromagnetic material. Under pressure of about 4 kbar or at the temperature of 318 K, a phase transition of the first kind from a ferromagnetic phase to a paramagnetic one, occurs in this material.

The samples of MnAs used in the experiments of references [1,2], had the form of a plate with linear sizes of $20 \times 20 \times 10$ mm and were made by compacting a powder with subsequent annealing. The experiments were carried out as follows [1,2]: the sample was put into a weak magnetic field B of about 100 Gs and exposed to the light flashes of a time duration about a few microseconds and with energy of about 500 J. On the sample there was reeled up a coil (henceforth we will call it a signal coil) connected directly to the oscilloscope (Fig. 1).

When the light flashed, in the signal coil, an electromotive force which was detected by the oscilloscope as current pulses was induced. As it was suggested in [1,2], the emergence of current pulses in a coil reeled up on the sample, very naturally could be explained by the magnetization changes occurring in manganese arsenide. The magnetization changes of MnAs (calculated by means of experimental data) are dependent on temperature and are depicted in Figure 2 (see also [2]). As it can be seen, the maximum change of magnetization is observed at $\sim T_c$ (318 K); however, the changes are also observed at temperatures considerably smaller than T_c. The authors took special care to cool the sample by the vapour of liquid nitrogen. This allowed them, as they believed, to exclude the thermal effect of light and to interpret the results of experiments as the observation of light-stimulated phase transition of MnAs from the ferromagnetic state to paramagnetic one at the temperature $T < T_c$. According to [1,2], the magnetization of MnAs-sample changes abruptly at such a transition and causes the change of magnetic flux threading the signal coil. As a result, electromotive force is induced in it and the corresponding current peaks are detected by the oscilloscope.

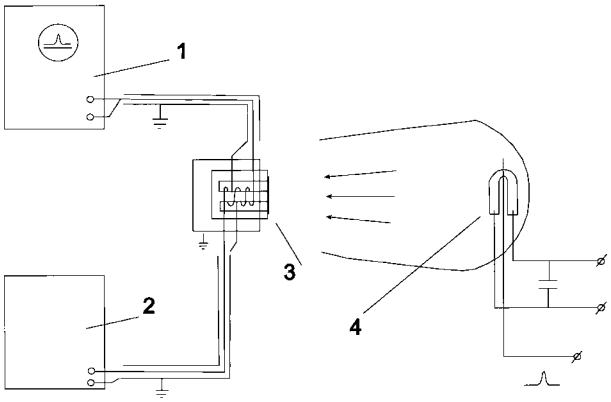

Fig. 1. Experimental setup of [1,2]: 1 - oscilloscope, 2 - energy supply, 3 - MnAs-sample set into external magnetic field, 4 - flash lamp.

However, the arguments put by the authors in favour of their interpretation of the observed phenomenon are, as we shall see, by no means convincing. To my mind, the results of these experiments have to, and can be

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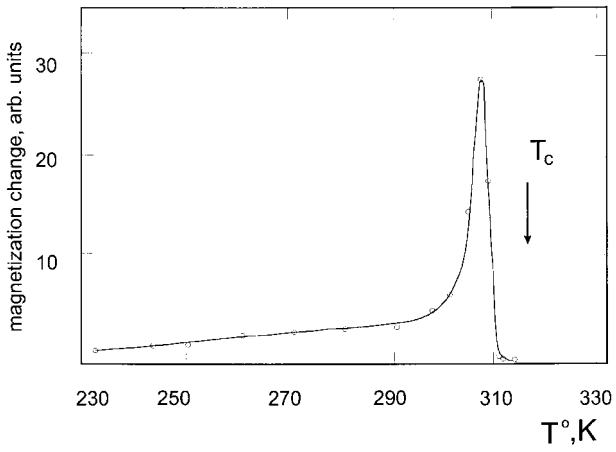

Fig. 2. Magnetization changes of MnAs *vs.* temperature.

explained in completely different way. Such an explanation makes up the content of this paper and to the author's mind can be instructive, especially in the light of recent investigations of an orbital diamagnetism due to persistent currents (see, for instance, Y. Ishikawa, H. Fukuyama, `cond-mat/9904052`).

2 Preliminary analysis

In reference [2] there was an attempt made to explain the ferromagnetic ordering in MnAs, as well as the results of experiments [1,2] in the framework of Ruderman-Kittel-Kasuya-Yosida (RKKY)-theory [3–5].

The PKKY-theory is based on the idea of indirect exchange coupling or so called $s-d(s-f)$ -exchange. According to this idea, all the electrons in a crystal are supposed to be divided into two groups. The electrons of the first group are the delocalized electrons of conduction band composed mainly of s-states. The electrons of another group are the localized electrons belonging to the partially occupied d- or f-states generically related to the $d(f)$-shell of magnetic ions of which the crystal is composed.

Supposing the single-electron approximation is valid, one can treat the $s-d(f)$ exchange model in terms of the Hamiltonian:

$$H = H_{\rm B} + H_{\rm M} + H_A, \tag{1}$$

where $H_{\rm B}$ is the Hamiltonian of conduction band electrons,

$$H_{\rm B} = \sum E_{\mathbf{k}} a^{\dagger}_{\mathbf{k}\sigma} a_{\mathbf{k}\sigma},$$

$a^{\dagger}_{\mathbf{k}\sigma}$, $a_{\mathbf{k}\sigma}$ are the creation and anihilation operators for the state $(\mathbf{k},\sigma)$, where $\mathbf{k},\sigma$ are the momentum and spin projection, respectively. $H_{\rm M}$ is the Hamiltonian of magnetic subsystem, which is usually assumed to be:

$$H_{\rm M} = \sum_{ik} J(\mathbf{i}-\mathbf{k})(\mathbf{S}_i \cdot \mathbf{S}_k) - g\mu_B \mathbf{B} \sum_i \mathbf{S}_i.$$

Here subscript i runs over all atoms, while subscript k is usually supposed to run over the nearest neighbours of an atom participating in the exchange interaction, $J(\mathbf{i}-\mathbf{k})$ is the energy of exchange interaction of localized spins which is often assumed to be constant, $\mathbf{S}_i, \mathbf{S}_k$ are the spin operators of paramagnetic ions, g is the Lande factor, $\mu_{\rm B}$ is Bohr magneton and $\mathbf{B}$ is an external magnetic field. The last term in (1), H_A, is the Ruderman-Kittel, or so called $s-d$ exchange which is responsible for the interaction between conduction electrons (c-electrons) and the electrons localized on magnetic ions. In one-band approximation it is of the form:

$$H_A = -A \sum (\mathbf{S}_i \cdot \mathbf{s})_{\sigma\sigma'} a^{\dagger}_{i\sigma'} a_{i\sigma}, \tag{2}$$

and the components of c-electron spin are

$$s^{\alpha}_g = \sum (s^{\alpha})_{\sigma\sigma'} a^{\dagger}_{g\sigma'} a_{g\sigma'},$$

where $a^{\dagger}_{g\sigma'}, a_{g\sigma'}$ are the creation and anihilation operators on the site g with the spin projection σ', s^{α}_g $(\alpha = x,y,z)$ are the following matrices:

$$s^x = \frac{1}{2}\begin{pmatrix} 0 & 1 \\ 0 & 1 \end{pmatrix}, \quad s^y = \frac{\rm i}{2}\begin{pmatrix} 0 & -1 \\ 0 & 1 \end{pmatrix}, \quad s^z = \frac{1}{2}\begin{pmatrix} 0 & 1 \\ 0 & -1 \end{pmatrix}.$$

The structure of (2) becomes more lucid, if we notice that in coordinate representation the exchange interaction between the c-electron on the $\mathbf{i}$-site and the electron of magnetic ion on $\mathbf{k}$- site is of the form: $[-A(\mathbf{i}-\mathbf{k})(\mathbf{S}_{\mathbf{k}} \cdot \mathbf{s})]$, where $\mathbf{s}$ is the spin operator of the c-electron. Since the exchange interaction is short range, one can restrict the consideration only to the nearest neighbours interaction. In this case the interaction energy $A(0)$ is of zeroth order with respect to the overlapping of Wannier function of c-electron and the wave function of localized electrons. If the electrons are on the nearest neighbour sites, the exchange energy is of the second order with respect to the overlapping of orbitals. As a result, $A(\mathbf{i}-\mathbf{k})$ is delta function-like, and if one deals with the indirect coupling, it is sufficient to take into account only the term $A(0) = A$, which actually appears in (2).

Thus, one can say that in the RKKY-model the magnetic ordering is, in fact, the ordering of localized spins just like in Heisenberg model; the contribution of c-electrons spin to total magnetic moment of the crystal is small compared with the spin of localized electrons, but just the c-electrons make the ordering of localized spins possible.

From the formal point of view, RKKY-theory is a sort of perturbation theory where the small perturbation factor $\lambda \ll 1$ is the ratio of exchange energy to the Fermi energy $\lambda \sim A/E_{\rm F}$. The parameter λ is small enough for some rare earth metals, some magnetic semiconductors, such as EuS and EuO, and some diluted alloys. It is commonly used to think that for such materials A is of a few tenths of eV and the Fermi energy is about 5 eV. One can use the Curie temperature $T_{\rm c}$ as the measure of exchange

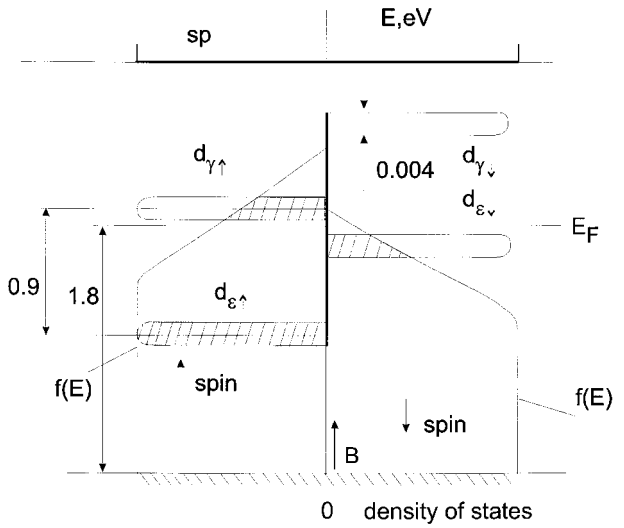

Fig. 3. Schematic band structure of MnAs.

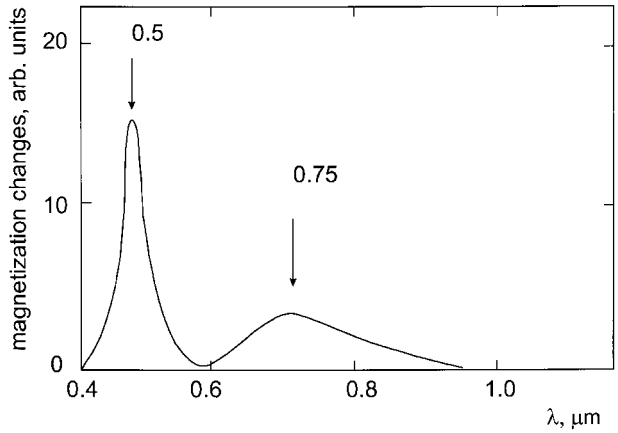

Fig. 4. Spectral dependence of the MnAs magnetization change.

interaction energy. That is why T_c for the materials described by RKKY-theory should be not greater than a few dozens of kelvins, since only then $\lambda < 1$.

Surely, it is not the case for manganese arsenide: for MnAs T_c is almost 20 times greater than for EuS and 5 times greater than for EuO, while the Fermi energy, in contrast, is smaller than that for EuS and EuO and is about 2 eV. As a result, the factor λ is not small and the RKKY-theory can hardly be applicable to the description of magnetic ordering in MnAs and for the interpretation of the experiments of reference [1,2]. This analysis makes us doubt of the validity of the hypothesis settled forth in [1,2], however attractive it might seem.

On the other hand, the experiments of references [1,2] unambiguously show that the electromotive force in the signal coil is really induced. It should be related somehow or other to the change of magnetic flux threading the coil, since it is hard to imagine howelse the matter be explained. Thus, the problem is to guess what really occurs in the sample of MnAs exposed to the light pulses and this is the topic of the next section.

3 New interpretation of experimental results

To begin with, let us analyse the MnAs-band structure with its relation to the experimental data reported in [2].

The conceptual band scheme for MnAs for the first time was proposed by Goodenough *et al.* [6]. It is based on the molecular orbital approach: the *s*- and *p*-states of Mn- and As-atoms are supposed to build up bonding and antibonding *sp*-bands; the bonding band (valence) is completely filled, while the antibonding band (conduction) is empty. In between there are the narrow subbands which are due to the overlapping of *d*-orbitals of Mn atoms. Subsequently, this scheme was elaborated by other authors; according to [7,8], the energy gap between the filled and empty bands is about 3.0 eV, the 3*d*-band is split by crystal field into very narrow spin-polarised $d_{\varepsilon\uparrow}, d_{\gamma\uparrow}$ and $d_{\varepsilon\downarrow}, d_{\gamma\downarrow}$ subbands (here $\uparrow$ and $\downarrow$ stand for spin-up and spin-down states) (Fig. 3). The energy gap between $d_{\varepsilon\uparrow}, d_{\gamma\uparrow}$ ($d_{\varepsilon\downarrow}, d_{\gamma\downarrow}$, resp.) is about 0.9 eV; the Fermi level is between $d_{\varepsilon\downarrow}$ and $d_{\gamma\uparrow}$- subbands at $T = 0$ K. The occupancy of each subband is determined by the Fermi-Dirac distribution function and the corresponding density of states; the population of $d_{\gamma\uparrow}$ tends to zero at the temperature $k_B T < E_g/2$.

The results of [2] clearly show (see Fig. 4) that there are two distinct maxima of MnAs magnetization changes with respect to the light wave length. The first one corresponds to $\lambda_1 = 0.49$ μm, while the other corresponds to $\lambda_2 = 0.75$ μm. So, the first maximum can be attributed to the dipole transition from *sp*-band to the empty $d_{\gamma\downarrow}$-subband, while the second one can be attributed to the transition to the partially occupied $d_{\varepsilon\downarrow}$-subband. Thus, to my mind the results of [1,2] can be explained as follows. Under the light pulses the population of empty $d_{\gamma\downarrow}$ and partially occupied $d_{\varepsilon\downarrow}$ subbands increases. The quasi-free electrons of these narrow subbands revolve about the direction of the small external magnetic field **B** during short time intervals proportional to the scattering time. Naturally, the orbital magnetic moment directed opposite to the external magnetic field corresponds to this circular motion. The emergence of an additional orbital magnetic moment leads to the changes of magnetic flux threading the signal coil. The last one, in its turn, causes the pulses of electromotive force detected by oscilloscope.

Put simply, to my mind, the authors of [1,2] observed the current pulses caused by the changes of magnetic flux which, in their turn were due to the *non-equilibrium light-induced diamagnetic moment* of the quasi-free electrons moving in the narrow *d*-subbands of the MnAs conduction band. That the *d*-subbands of MnAs are narrow, is very important, since the narrower the subband is, the larger is the electron scattering time and if the conduction band is sufficiently narrow, the electrons have enough of time to revolve several times about the external magnetic field, producing an orbital magnetic moment. In the case of a wide conduction band the scattering time is too small for

the electron could make several revolutions and hence, no orbital momentum arises.

Now let us analyse quantitatively the proposed model. Let the Hamiltonian of an electron of *sp*-band interacting with classical electromagnetic field of an incident light be

$$\hat{H}(t) = \hat{H}_0 + \hat{H}_1(t).$$

Here $\hat{H}_1(t) = -\mathbf{dE}(\mathrm{t})$ describes the interaction of the electron with a classical electromagnetic field in the dipole approximation, $\mathbf{d}$ is the dipole transition operator (remember, we attributed the maxima of magnetization changes to the $sp \rightarrow d_{\gamma\downarrow(\varepsilon\downarrow)}$ transitions; the last one corresponds to $\Delta L = \pm 1$ selection rule,where L is the angular momentum quantum number); $\mathbf{E}(\mathrm{t})$ is the electric field of the electromagnetic wave.

The external magnetic field $\mathbf{B}$ is supposed to be included in $\hat{H}_0$; we also assume the electrons to interact with the crystal lattice vibrations (phonons) and take into account the relaxation processes simply by introducing a phenomenological relaxation time τ. We will ignore the electron-electron interaction altogether assuming it to be inessential.

Thus, the evolution of the quantum subsystem (electrons) can be treated in terms of von Neumann equation for the density matrix $\rho(t)$:

$$\mathrm{i}\hbar\frac{\partial\rho}{\partial t} = \hat{H}\rho - \rho\hat{H} - \mathrm{i}\frac{\hbar}{\tau}(\rho - \rho^{(0)}),$$

where $\rho^{(0)} = \rho(t = -\infty)$ is the density matrix for the steady state.

Since in the experiments of references [1,2] a flash lamp was used rather than a laser as the source of light, it is natural to expand the density matrix into a series: $\rho = \rho^{(0)} + \rho^{(1)} + ...$ and restrict consideration only to the linear approximation. We arrive then at the next linearised equation for the density matrix:

$$\mathrm{i}\hbar\frac{\partial\rho^{(1)}}{\partial t} = H_1\rho^{(0)} - \rho^{(0)}H_1 + H_0\rho^{(1)} - \rho^{(0)}H_0 - \mathrm{i}\frac{\hbar}{\tau}\rho^{(1)}. \tag{3}$$

Let it be $|k\rangle$- the complete orthonormal set of eigenvectors of H_0, that is

$$(\hat{H}_0 - \varepsilon_k)|k\rangle = 0,\ \langle l|k\rangle = \delta_{lk}.$$

Taking into account that in energy representation, the steady state density matrix $\rho^{(0)}$ is of the form

$$\langle s|\rho^{(0)}|l\rangle = \frac{\mathrm{e}^{-\beta\varepsilon_l}\langle\varepsilon_s|\varepsilon_l\rangle}{\sum_l \mathrm{e}^{-\beta\varepsilon_l}} = f_0(\varepsilon_l)\delta_{sl},$$

where $f_0(\varepsilon_l)$ is the distribution function, one can rewrite equation (3) in the form

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\rho^{(0)}_{kl}(t) = [f_0(\varepsilon_l) - f_0(\varepsilon_k)]H^{(1)}_{kl} + (\varepsilon_k - \varepsilon_l)\rho^{(1)}_{kl}(t) - (\mathrm{i}\hbar/\tau)\rho^{(1)}_{kl}(t). \tag{4}$$

If one searches for the solution to (4) in the form

$$\rho^{(1)}_{kl}(t) = \int_{-\infty}^{\infty}\rho^{(1)}_{kl}(\omega)\mathrm{e}^{-\mathrm{i}\omega t}\mathrm{d}\omega,$$

then one arrives at

$$\rho^{(1)}_{kl}(\omega) = \frac{[f_0(\varepsilon_l) - f_0(\varepsilon_k)]d_{kl}E(\omega)}{\varepsilon_k - \varepsilon_l - \hbar\omega - \mathrm{i}\hbar\eta},$$

where $d_{kl} = e\langle k|\mathbf{r}|l\rangle$ is the matrix element of the dipole transition operator, $\eta = \tau^{-1}$ and $E(\omega)$ is the Fourier transform of the classically treated electromagnetic field of light emitted by the flash lamp:

$$E(t) = \int_{-\infty}^{\infty}E(\omega)\mathrm{e}^{(\mathrm{i}\omega t)}\mathrm{d}\omega.$$

The mean value of the z-component of induced magnetic moment (the z-axis is assumed to coincide with the direction of the external magnetic field) can be calculated in the framework of the *Kubo linear response approach* [9]. We get:

$$\langle\mathcal{M}_z\rangle_t = \sum_{k,l}\int_{-\infty}^{\infty}\frac{[f_0(\varepsilon_l) - f_0(\varepsilon_k)]d_{kl}(\hat{\mathcal{M}}_z)_{kl}E(\omega)\mathrm{e}^{\mathrm{i}\omega t}\mathrm{d}\omega}{\varepsilon_k - \varepsilon_l - \hbar\omega - \mathrm{i}\hbar\eta}, \tag{5}$$

where $(\hat{\mathcal{M}}_z)_{kl} = \langle k|\mathcal{M}_z|l\rangle$ is the matrix element of the z-component of the orbital magnetic moment operator.

Let us remember firstly, that the time scale of magnetization changes observed in [1,2] was about pulse duration and secondly, that a flash lamp was used as the source of light. Hence, in considering the classical electromagnetic field $E(t)$ one should use rather the so called *slowly varying envelope approximation* [10]. Thus, the shape of the light pulses used in [1,2] to a good accuracy can be approximated by a rectangular one:

$$E(t) = \begin{cases} E_0, & 0 \le t \le t_0, \\ 0, & t < 0,\ t > t_0, \end{cases}$$

while its Fourier transform is of the form: $E(\omega) = (\mathrm{i}E_0/\omega)(1 - \exp(\mathrm{i}\omega t_0))$. Using the last formula, one can easily calculate the integral in (5). Indeed, let us consider an integral

$$I = \int_{-\infty}^{\infty}\frac{\mathrm{i}E_0(1 - \mathrm{e}^{\mathrm{i}\omega t_0})\mathrm{e}^{-\omega t}}{\omega(\varepsilon_k - \varepsilon_l - \hbar\omega - \mathrm{i}\hbar\eta)}\mathrm{d}\omega \tag{6}$$

and make an analytical continuation of the integrand over the lower half-plane $\Im\,\omega < 0$ (here $\Im$ stands for the imaginary part of the subsequent expression). It is easily seen that now the integrand obeys the *Jordan Lemma* [11] and hence, $I = -2\pi\mathrm{i}Res[F(\omega)\mathrm{e}^{-\mathrm{i}\omega t}]$, where

$$F(\omega) = \frac{\mathrm{i}E_0(1 - \mathrm{e}^{\mathrm{i}\omega t_0})}{\omega(\varepsilon_k - \varepsilon_l - \hbar\omega - \mathrm{i}\hbar\eta)},$$

and $\hbar^{-1}(\varepsilon_k - \varepsilon_l) - \mathrm{i}\eta = \omega_{kl} - \mathrm{i}\eta = z_0$ is the only pole of the integrand in the lower half-plane $\Im\,\omega < 0$. As a result we have:

$$\langle \mathcal{M}_z \rangle_t = -2\pi \sum_{k,l} \frac{[f_0(\varepsilon_l) - f_0(\varepsilon_k)] d_{kl} (\mathcal{M}_z)_{kl} E_0}{\hbar(\omega_{kl} - \mathrm{i}\eta)} \times \left[\mathrm{e}^{-\eta t} \mathrm{e}^{-\omega_{kl} t} - \mathrm{e}^{\mathrm{i}\omega_{kl}(t_0 - t)} \mathrm{e}^{\eta(t_0 - t)} \right].$$

Now it is easily seen that, indeed, induced magnetic moment and hence, the magnetization changes could be observable only for the time interval $t \sim t_0$, because for the time $t > t_0$ they decrease exponentially.

In order to discuss the spectral dependence of MnAs magnetization changes, one should analyze the Fourier component of induced magnetic moment:

$$(\mathcal{M}_z)_\omega = \sum_{k,l} \frac{[f_0(\varepsilon_l) - f_0(\varepsilon_k)] d_{kl} (\mathcal{M}_z)_{kl}}{\hbar(\omega_{kl} - \omega - \mathrm{i}\eta)} E(\omega), \qquad (7)$$

and its real part, which is of the form

$$\Re(\mathcal{M}_z)_\omega = -\sum_{k,l} \frac{[f_0(\varepsilon_l) - f_0(\varepsilon_k)] d_{kl} (\mathcal{M}_z)_{kl} E_0 (\Delta_{kl} \sin\omega t - \eta(1 - \cos\omega t_0))}{\hbar\omega(\Delta_{kl}^2 - \eta^2)}, \qquad (8)$$

where $\Delta_{kl} = \omega_{kl} - \omega$ and $\Re$ stands for the real part of the subsequent expression.

Here it is necessary to make some comments concerning subsequent calculations. The dynamic of electrons in an energy band and external fields can be described by an equation of the form:

$$\left[\varepsilon_n + \frac{(\hbar\nabla + (e/c)\mathbf{A})^2}{2m^*} + U(\mathbf{r}) \right] \Psi(\mathbf{r}) = \mathrm{E}\Psi(\mathbf{r}) \qquad (9)$$

where ε_n is the energy of corresponding band, $\mathbf{A}$ is the vector potential, m^* is the effective mass and $U(\mathbf{r})$ is the potential arising due to space-charge or any other discontinuity or, for instance, other confining potential etc. Although equation (9) looks just like the Schrödinger equation, it is really what is called a single band effective mass approximation. The lattice potential, which is periodic on atomic scale, does not appear explicitly in equation (9); its effect is incorporated through the effective mass m^* which we will assume to be spatially constant. It should be noted also that the wave function one calculates from equation (9) is not the true wave function but its smoothed out version that does not show any rapid variations on the atomic scale.

Since $U(\mathbf{r})$ is often referred to as the confinement potential responsible for the size quantization in micro- and mesoscopic structures (see, for example, [12]), we can suppose it to be zero, because we deal with a macroscopic sample.

If the vector potential to be $\mathbf{A} = (-(1/2y)B, (1/2x)B, 0)$, then the Hamiltonian of an electron in a uniform magnetic field is:

$$H_0 = \frac{1}{2m^*} \left[\left(p_x - \frac{e}{2} y B \right)^2 + \left(p_y + \frac{e}{2} x B \right)^2 + p_z \right]. \qquad (10)$$

Using Hamiltonian (10), after a round of tiresome calculations (see Appendix), one arrives at

$$\Re(\mathcal{M}_z)_\omega = -\frac{\sqrt{2}\pi\hbar e^2 a_\mathrm{B} E_0 (\mathbf{e}_\rho \mathbf{n}_\mathbf{E})}{m^* \hbar\omega(\Delta_{sd}^2 - \eta^2)} \times (\Delta_{sd} \sin\omega t_0 - \eta(1 - \cos\omega t_0)) F(f(\varepsilon_d), f(\varepsilon_s), n_\rho, m), \qquad (11)$$

where $F(f(\varepsilon_d), f(\varepsilon_s), n_\rho, m)$ is a very complicated function of its arguments (n_ρ, m are the corresponding quantum numbers), explicit form of which is given in the Appendix; $\Delta_{sd} = \hbar^{-1}(\varepsilon_d - \varepsilon_s - \hbar\omega)$ is the detuning, s and d stand for s-and d-bands respectively, $a_\mathrm{B} = \sqrt{\hbar/|e|B}$ is the magnetic length.

One should notice that in accordance with (11), the dimension of the Fourier component of the induced magnetic moment is equal to (*magnetic moment/frequency*). However, to make reasonable estimates which could be compared with experimental data, one should deal with the mean value of the spectral component of the induced magnetic moment rather than the Fourier component of the magnetic moment. Since in [1,2] a flash lamp rather than a laser was used as the source of light, one can suppose the detuning Δ_{sd} to be much greater than η. Thus, averaging $\Re(\mathcal{M}_z)$ over a small frequency interval $\Delta\omega$, one can substitute $\cos\omega t_0$ and $\sin\omega t_0$ by their mean value of 0 and get the next formula for the mean value of the induced magnetic moment:

$$\langle \mathcal{M}_z \rangle \sim -\frac{\sqrt{2}\pi\hbar e^2 a_\mathrm{B} \langle E_0 \rangle \langle \mathbf{e}_\rho \mathbf{n}_\mathbf{E} \rangle \eta \Delta\omega}{m^* \hbar\omega \Delta_{sd}^2} \times F(f(\varepsilon_d), f(\varepsilon_s), n_\rho, m).$$

Also it is necessary to add here some comments concerning firstly, the meaning of the quantity: $\langle E_0 \rangle \langle \mathbf{e}_\rho \mathbf{n}_\mathbf{E} \rangle$ and second, the relation between the induced magnetic moment and the changes of magnetic flux threading the signal coil. Actually, in the experiments discussed, the measurable quantity was the intensity of light, but not E_0. Hence, the last one should be expressed in terms of averaged power flux: $\langle W \rangle = 1/2(\varepsilon_0 c |\langle E \rangle|^2)$, where ε_0 is the dielectric constant and c is the light velocity.

As for the relation between the induced magnetic moment and the changes of magnetic flux, let us notice that the dimension of the magnetic moment is $[\mathcal{M}] = L^{5/2} M^{1/2} T^{-1}$, where L, M, T stand for the units of length, mass and time respectively. In order to construct, by means of $\mathcal{M}$, the quantity of the dimension of magnetic field $[B] = L^{-1/2} M^{1/2} T^{-1}$, one should multiply $\mathcal{M}$ by the quantity which is of L^3 dimension. The only relevant quantity of such dimension is the volume V_0 of the part of the sample where the light is absorbed. Now the change of magnetic flux can be estimated as $-\mathrm{d}\Phi/\mathrm{d}t \approx -\langle \mathcal{M}_z \rangle V_0 / t_0$.

Now we are well equipped to proceed to the numerical estimates. To this end, let us have the data of [1,2] power flux incident on the sample is about 1 W cm^{-2}, $t_0 \sim 10^{-5}$ s, sample area is of 20×10 mm, $B = 10^{-2}$ T, thickness of the layer where the incident light is absorbed, is of 50 nm; the energy gap between *sp*-and $d_{\gamma\downarrow}$- bands is about 2.5 eV, the width of $d_{\gamma\downarrow}$- band (and the spectral range $\Delta\omega$) is of 0.004 eV. Above all, we suppose $\tau \sim \omega_{\rm B}^{-1} = |e|B/m^* \sim 2 \times 10^{-9}$ s and $m^* \approx m_0$. Then, even if one assumes the number of loops of the coil to be $n = 1$ (the exact number was not reported in [1,2], but certainly it cannot be less than 1), the electromotive force calculated in this way would be about 100 μV. No doubt such signal is quite well to be detected by the standard oscilloscope even without intermediate amplification. However, the calculations made in such way as to underestimate the effect and the reason for that is the following. We estimated the magnetic flux change by the formula $-{\rm d}\Phi/{\rm d}t \sim \Delta\Phi/t_0$, while in fact, instead of flash duration t_0, one should rather use the rates of flash rise and flash decay. These can be much shorter than t_0; unfortunately, the rates of pulse rise and pulse decay also were not reported in [1,2].

4 Discussion and conclusions

Proceeding to the general discussion, it seems worthy to add at first some comments on the problem of light-induced phase transitions. Apart from trivial heating, there are other ways light can affect the phase transition in the solid state. Sometimes, contrary to the heating, these mechanisms can increase the ordering. For instance, at a given temperature, the magnetic ordering can be greater under the effect of light than without it. However, dealing with these transitions, one has to be careful when interpreting obtained results, because the basic difference between the usual phase transition and the light-induced transition is that the latter occurs under non-equilibrium conditions. As a result, for their description one cannot use the thermodynamic potentials such as, free energy. In thermal equilibrium the probability measure can in principle be expressed through an appropriate ensemble. For non-equilibrium systems an equally powerful concept is missing. However, the different tools for the treatment of non-equilibrium systems were already developed, among them the grand canonical partition function (see [13] and the references cited therein) and the entropy generation rate [14].

Nevertheless, there were already reported reliable experimental data [15] which could be interpreted as the observation of light-induced phase transitions in a semiconductor from the paramagnetic state to a ferromagnetic one. The basic idea here is that during light absorption the *helicity* (quantum number which is the projection of particle's spin onto the particle's wave vector) is conserved. It means that using circularly polarized light one can rise up the ordering of spins by means of their optical orientation. Such optical orientation of spins was indeed observed in an EuS-magnetic semiconductor [15]. The authors of reference [15] used a circularly polarized laser beam together with additional modulation, since, as they have shown, unmodulated unpolarized light led mainly to the heating of the sample. However, the shift of $T_{\rm c}$ observed for light-induced phase transitions was only about 0.1 K, not 10–20 K as was reported in [1,2].

Thus, our critical analysis shows that the explanation of the observed light-induced magnetization changes of MnAs proposed in [1,2] and based on the hypothesis of light-induced phase transition, is far to be satisfactory. Here instead, we propose completely different approach which, in fact, suggests that at least afar off the vicinity of $T_{\rm c}$, there are no real magnetization changes as in case of phase transition, but there are the changes of magnetic flux threading the MnAs-sample. These changes are due to *non-equilibrium light-induced diamagnetic moment of the quasi-free electrons of d-subbands of the* MnAs-*conduction band*. As for the nearest vicinity of $T_{\rm c}$, no doubt there is a phase transition here, but an ordinary thermodynamic phase transition caused by the thermal effect of light, that is, by heating of the sample.

As for the last remark in the previous section concerning the necessity to take into account time duration of the flash (pulse) rise and flash (pulse) decay, it suggests also that in fact, there should be observed *two current pulses*, the first one induced by the flash *rise* (or the *front edge* of the pulse) and the second induced by the flash *decay* (or the *back edge* of the pulse). Curiously enough, in the experiments of [1,2] there were *two pulses detected* , but the authors treated it as the argument in favour of their hypothesis of the light-induced phase transition from the ferromagnetic state to a paramagnetic one at the temperature $T < T_{\rm c}$, since as they wrote, *if it were the ordinary phase transition, the back transition from paramagnetic state to ferromagnetic state would occur after elapse of time much greater than the flash duration.* To the author's mind, it is just a misinterpretation, whereas the natural explanation of these results is proposed above.

In the framework of proposed model, the difference of the maxima's heights in Figure 4 also can be easily understood. Remember, the first maximum at $\lambda = 0.49$ μm corresponds to the transition from the valence band to the empty $d_{\gamma\downarrow}$ subband, while the second one corresponds to the transition to the partially occupied $d_{\varepsilon\downarrow}$ subband. Thus, in accordance with formulae (7) and (8), the induced magnetic moment should be greater for the first transition.

The experimental verification of the present explanation of the observed effect, is simple. First, it is sufficient to switch off the magnetic field: if the explanation proposed here is correct, no current pulses should be observed in the signal coil. Secondly, one could increase the current pulses by increasing the rates of flash rise and flash decay. By decreasing these rates (*i.e.* expanding the front and back edges of light flash) one could decrease the current pulses. Thirdly, by increasing time duration of the flash and increasing simultaneously the rates of its rise and decay, one could also increase the time interval between the

two current pulses. This last idea suggests that, in spite of doubtful interpretation, the effect observed in [1,2] is a very interesting one and as such could be a useful experimental technique, especially if the flash lamp would be substituted by laser. This technique could be used, for instance, for the investigation of the mechanisms of electron scattering in magnetic materials of band structure similar to that of MnAs.

As the final remark, perhaps it is worth mentioning, that the effect considered here resembles to some extent orbital diamagnetism in mesoscopic systems [16]. Such systems exhibit another type of electric current, which is without dissipation, in contrast to the transport currents. These are so called persistent currents, *i.e.*, orbital currents induced in micro- and mesoscopic structures by the external magnetic field [17] and they are the sources of orbital diamagnetism. It is clear that in our case the "orbital currents", that is, the revolutions of electrons in narrow *d*-subbands, though not persistent, are nevertheless long-lasting, since the scattering in narrow *d*-subbands is weakened and as a consequence, the orbital diamagnetism arises which is also not "persistent" but light-induced and non-equilibrium.

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Appendix

Here we derive the formula (11) for the real part of the Fourier component of induced magnetic moment.

Starting with the Hamiltonian (10) and introducing the cylindrical coordinates (ρ, φ, z), one can write down the solution to the corresponding Schrödinger equation in the form:

$$\Psi_{n_\rho,m}(\rho,\varphi,z) = \frac{\mathrm{e}^{\mathrm{i}m\varphi}}{\sqrt{2\pi}}\mathrm{e}^{\mathrm{i}p_z/h}R_{n_\rho,m}(\rho),$$

$$n_\rho = 1,2,...; m = 0,\pm1,...,\pm n_\rho,$$

$$R_{n_\rho,m}(\rho)C\exp[-\frac{1}{2}(\frac{\rho^2}{2a_{\mathrm{B}}^2})]\rho^{|m|}\Phi(-n_\rho,|m|+1,\frac{\rho^2}{2a_{\mathrm{B}}^2}),$$

here C is the normalizing constant, $\Phi(-n_\rho,|m|+1,\rho^2/2a_B^2)$ is the degenerate hypergeometric function.

By means of the last formulae, the matrix element of the z-component of induced magnetic moment can be represented as follows ($\{s\}$ stands for the set of quantum numbers $\{n_\rho, m\}$):

$$\langle s^{'}|\mathcal{M}_z|s\rangle = -\frac{em}{2m^*}\delta(p_z^{'}-p_z)\delta_{mm'} \times \int_0^\infty R_{n_\rho^{'},m'}(\rho)R_{n_\rho,m}(\rho)\rho\mathrm{d}\rho,$$

(remember, m^* stands for the effective mass while m is the quantum number). Substituting in the last integral $\rho^2/2a_{\mathrm{B}}^2$ by x, using the relation between degenerate hypergeometric function and the Laguerre polinomials [18]:

$$\Phi(-n_\rho,|m|+1,x) = \frac{n_\rho!|m|!}{(n_\rho+|m|)!}L_{n_\rho}^{|m|}(x),$$

and taking into account that

$$\int_0^\infty \mathrm{e}^{-x}x^\alpha L_{m'}^\alpha(x)L_m^\alpha(x)\mathrm{d}x = \begin{cases} 0,\ m\neq m^{'},\ \mathrm{Re}\alpha>-1, \\ \Gamma(\alpha+m^{'}+1)/m^{'}!,\ m=m^{'},\ \mathrm{Re}\alpha>0, \end{cases}$$

one gets

$$(\mathcal{M}_z)_{s's} = -\frac{m\hbar e}{2m^*}\delta(p_z^{'}-p_z).$$

Here also the natural normalizing condition

$$\int_0^\infty \left[R_{n_\rho m}(\rho)\right]^2\rho\mathrm{d}\rho = 1$$

was used. The matrix element of the dipole momentum operator in the cylindrical coordinates is of the form:

$$r_{s's} = e\left\{\langle s^{'}|\rho|s\rangle(\mathbf{e}_\rho\cdot\mathbf{n_E}) + \langle\mathbf{s}^{'}|\mathbf{z}|\mathbf{s}\rangle(\mathbf{e}_\mathrm{z}\cdot\mathbf{n_E})\right\},$$
$$\mathbf{n_E} = \frac{\mathbf{E}}{|\mathbf{E}|},$$

and $\mathbf{e}_\rho, \mathbf{e}_\mathrm{z}$ are the unit vectors of ρ- and z-axes, respectively. Taking into account that in equations (7, 8) summation includes also the integration over the components of the momentum p_z, one gets the next expression for the spectral component of the induced magnetic moment projection:

$$\Re\langle\mathcal{M}_z\rangle_\omega = -\frac{\pi\sqrt{2}\hbar e^2a_{\mathrm{B}}E_0(\mathbf{e}_\rho\cdot\mathbf{n}_E)(\Delta_{sd}\sin\omega t_0-\eta(1-\cos\omega t_0))}{m^*\hbar\omega(\Delta_{sd}^2-\eta^2)} \times\sum_{n_\rho m}\left[f_0(\varepsilon_d,n_\rho,m)-f_0(\varepsilon_s,n_\rho,m)\right]\frac{m\Gamma(3/2+|m|)}{n_\rho!|m|!} \times\left(\sum_k^{n_\rho}C_{n_\rho}^k\left[{}_2F_1^{(n_\rho-k)}(\alpha,\beta,\gamma;w(\eta))y^k(\eta)\right]_{\eta=0}\right),$$

where $\alpha = |m|/2+3/4; \beta = |m|/2+5/4; \gamma = |m|+1; w(\eta) = 16\eta^2/(1-\eta^2)^2$; ${}_2F_1^{(n_\rho-k)}(\alpha,\beta,\gamma;w(\eta))$ is the Gauss hypergeometric function; $y(\eta) = (1-\eta)^{1/2}(1+\eta^{-(|m|+3/2)}), C_{n_\rho}^k$ stands for the binomial coefficients ("choice numbers", $C_{n_\rho}^k = \binom{n_\rho}{k}$) that is, the formula (11), where

$$F(f(\varepsilon_d),f(\varepsilon_s),n_\rho,m) = \sum_{n_\rho m}\left[f_0(\varepsilon_d,n_\rho,m)-f_0(\varepsilon_s,n_\rho,m)\right]\frac{m\Gamma(3/2+|m|)}{n_\rho!|m|!} \times\left(\sum_k^{n_\rho}C_{n_\rho}^k\left[{}_2F_1^{(n_\rho-k)}(\alpha,\beta,\gamma;w(\eta))y^k(\eta)\right]_{\eta=0}\right).$$

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