THEORY OF THE RAMAN EFFECT DUE TO THE MAGNON-SPIN FLIP EXCITATION IN ANTIFERROMAGNETIC SEMICONDUCTORS

By A. Stasch

Institute of Physics, Technical University, Poznań*

(Received February 10, 1976)

In this paper estimates are given for the processes by which the s–d exchange interaction may contribute to the Raman scattering due to the magnon-spin flip of the conduction electron processes in the antiferromagnetic semiconductors. It is shown that for the typical value of the s–d exchange integral, i. e. 0.01 to 0.1 eV, the extinction coefficient in the antiferromagnetic semiconductors can be larger for two orders of the value than that in the ferromagnetic materials. The value of the extinction coefficient in the antiferromagnetic semiconductors is found to be $10^{-9}-10^{-10}$ cm^{-1} sr^{-1}. It is also shown that the change of the value of an absorbed energy in an external magnetic field allows us to determine the gyromagnetic factor for the conduction electrons.

1. Introduction

Recently interesting observations as well as the theoretical predictions have been reported on the Raman processes related with magnons in magnetic insulators [1–4] and the spin flip excitations in the nonmagnetic semiconductors [5–8].

On the other hand in the actually available literature [9–11] there occurs a lack of observations and interpretation of the phenomena due to the interaction between the conduction electrons and the magnetic structure of the localized spins. The purpose of the present paper is to survey the theories of the Raman processes by using the s–d exchange model of the antiferromagnetic semiconductor.

2. The Hamiltonian of an antiferromagnetic semiconductor interacting with photons

The Hamiltonian for a magnetic semiconductor with the s–d exchange interactions and the applied homogeneous constant magnetic field can be written in the following from [12–14]

$$H = H_e + H_m + H_{s-d} + H_{e-r}. \tag{2.1}$$

* Address: Instytut Fizyki, Politechnika Poznańska, Piotrowo 3, 60-965 Poznań, Poland.

The operator H_e describing the system of the unperturbed electrons of the conduction and valence band has the following form

$$H_e = \sum_{k,m,s} E_{k,m,s} c^{+}_{k,m,s} c_{k,m,s}, \tag{2.2}$$

where $c_{k,m,s}$, $c^{+}_{k,m,s}$ denote the annihilation and creation operators of an electron with the wave vector $\boldsymbol{k}$ and the spin s in the band m.

The value of the energy $E_{k,m,s}$ is defined as follows

$$E_{k,c,s} = E_{k,c} + s g_c H_z, \tag{2.3a}$$

$$E_{k,v,s} = E_{k,v}, \tag{2.3b}$$

where $E_{k,m}$ is the energy of an electron with the wave vector $\boldsymbol{k}$ in the Bloch state of the m-th band under the absence of an external magnetic field.

It should be noted that for the valence electrons a part of their energy dependent upon the external magnetic field was induced to the term H_m which takes the following form [1]

$$H = \sum_{q} E_{1q} f^{+}_{1q} f_{1q} + E_{2q} f^{+}_{2q} f_{2q}, \tag{2.4}$$

where $f^{+}_{1q}, f^{+}_{2q}, f_{1q}, f_{2q}$ are the creation and annihilation operators of the magnons in the first or second magnetic sublattice, respectively; whereas the magnon energies E_{1q} and E_{2q} are defined by the following expressions [1]

$$E_{1q} = E_q + g_v |\mu_B| H_z, \tag{2.4a}$$

$$E_{2q} = E_q - g_v |\mu_B| H_z, \tag{2.4b}$$

with

$$E_q = g_v \mu_B H_E \left[\left(1 + \frac{H_A}{H_E}\right)^2 - D_q^2 \right]^{1/2}, \tag{2.4c}$$

$$D_q = Z^{-1} \sum_{\langle m \rangle} e^{iq(R_n - R_m)}. \tag{2.4d}$$

The symbol $\langle m \rangle$ at the summation denotes that the sum is limited to the nearest neighbours of the spin S_n at the site R_n. H_A and H_E denote the anisotropic field and the exchange field, respectively [1].

The idea of an exchange interaction between the conduction electrons and localized spins sketched qualitatively by Vonsovsky [14] and then worked out mathematically by Kasuya [15], was applied in interpreting the transport phenomena in semiconductors possesing magnetic structures by many authors [16, 17].

The consideration based on the perturbation calculation leads to the following form of the operator H_{s-d} [14, 15]

$$H_{s-d} = -\left(\frac{S}{N}\right)^{1/2} \sum_{k',k,q} J_{s-d}(\mathbf{k}, \mathbf{k}')\,(v_q+u_q)$$

$$\times [c^{+}_{k,c,+}c_{k',c,-}(f^{+}_{2q}-f_{1q})\delta_{k',k+q}+c^{+}_{k,c,-}c_{k',c,+}(f^{+}_{1q}+f_{2q})\delta_{k',k-q}]$$

$$+\frac{1}{N}\sum_{k,q} J(\mathbf{k}, \mathbf{k})\,(c^{+}_{k,c,-}c_{k,c,-}-c^{+}_{k,c,+}c_{k,c,+})\,(f^{+}_{1q}f_{1q}-f^{+}_{2q}f_{2q}), \tag{2.5}$$

where [1]

$$u_q = \cosh\frac{A_q}{2}, \tag{2.5a}$$

$$v_q = \sinh\frac{A_q}{2}, \tag{2.5b}$$

$$\tanh A_q = \frac{-D_q}{\left(1+\dfrac{H_A}{H_E}\right)} \tag{2.5c}$$

and $J(\mathbf{k}, \mathbf{k})$ denotes the Fourier transformation of the s–d exchange integral [13, 14].

The last term of the Hamiltonian (1.1) describing the interaction between the electron system and the electromagnetic field can be rewritten in the following form [7, 18]:

$$H_{e-r} = \left(\frac{2\pi e^2\hbar}{m^2\varepsilon V}\right)^{1/2} \sum_{l,k,s} \frac{1}{(\omega_l)^{1/2}}\,u_l$$

$$\times [p_{k+l,m;k,m'}c^{+}_{k+l,m,s}c_{k,m,s}a_l+p_{k,m;k,m'}c^{+}_{k-l,m,s}c_{k,m,s}a^{+}_l)], \tag{2.6}$$

where

$$p_{k,m;k,m'} = \langle \mathbf{k}, m, s|p|\mathbf{k}, m', s\rangle$$

denote the matrix elements of the momentum operator calculated in the Bloch function system $|\mathbf{k}, m, s\rangle$. a^{+}_l, and a_l are the creation and annihilation operators of photons with the wave vector l the energy and the polarization u_l, respectively. After having described the Hamiltonian of the system to the second-quantization representation we can proceed to discuss the Raman light scattering on magnetic elementary excitations in antiferromagnetic semiconductors.

3. The Raman process connected with the generation of an elementary excitation of the type: the magnon-spin flip of the conduction electron

In our paper we will show that in the magnetic semiconductors we should expect the occurrence of the Raman effect connected with the generation of the new type two--particle elementary excitation consisting of the magnon and spin flip of the conduction electron. In the connection with the necessity of the annihilation of the scattered photon and the excitation of the type of the magnon-spin flip of a conduction electron, the non--zero contribution to the probability $W(t)$ of the occurrence of such a process until the moment t is obtained in the following form [19]:

$$W(t) = \sum_{l_2 k, k', q} \Bigg| \sum_{c,d} \frac{\langle n_{l_1}-1;\ n_{l_2}+1;\ n_{q_2}+1;\ n_{k,c,+} = 1;\ n_{k',c,-} = 0 |\, H | c\rangle}{(E_c - E_0)(E_d - E_0)}$$

$$\times \langle c|H'|d\rangle \langle d|H'|n_{k,c,+} = 0;\ n_{k',c,-} = 1;\ n_{q_2}, n_{l_1}, n_{l_2}\rangle \Bigg|^2$$

$$\times \delta\left(\hbar\omega_{1} - \hbar\omega_{l_2} - E_{2q} + \frac{E_{k,c,-} - E_{k,c,+}}{\hbar}\right) \tag{3.1}$$

where $H' = H_{s-d} + H_{e-r}$. E_0 and E_c, E_d denote the values of the energy in the initial and intermediate states, respectively. $|c\rangle$, $|d\rangle$ are the unperturbed states of the system under consideration and n_{l_1}, n_{l_2}, n_q, $n_{k,c,+}$, $n_{k,c,-}$, denote the occupation numbers for the photons incident and scattered, the magnons with the wave vector $\boldsymbol{q}$ and the conduction electrons in the Bloch states $|\boldsymbol{k}, c, +\rangle$, $|\boldsymbol{k}, c, -\rangle$, respectively. The quantities $\hbar\omega_{l_1}$, $\hbar\omega_{l_2}$, E_{2q}, $E_{k,c,-}$, $E_{k,c,+}$ are the energies of these particles ordered in the analogical way.

In our further considerations, similarly like other authors [1–7], we will restrict ourselves to the spontaneous Raman effect and we will consequently lead the calculations at the low temperature region; we can then assume that in the initial state n_{l_2}, and n_q are equal to zero.

Moreover, in order to simplify the calculations we will take into account the strong magnetic polarization of the conduction band via the external magnetic field. In consequence of such polarization, at small concentrations of the conduction electrons, the states of the band with the spin $s = +\frac{1}{2}$ are occupied and those with $s = -\frac{1}{2}$ are empty. The scheme of occupation of the conduction band is given in Fig. 1. The keen analysis of the scattering processes shows that the contribution to $W(t)$ is given due to the scattering processes shown in Fig. 2. A typical example of the products of the matrix elements, corresponding to the scheme in Fig. 2, can be analytically written in the following form:

$$\frac{u_{l_1} p_{k-l_2,v;k',c} J_{s-d}(\boldsymbol{k};\ \boldsymbol{k}'+\boldsymbol{q}) u_{l_2} p_{k+q,c;k-l_2,v}}{(E_{k,c,-} - E_{k'+q-l_1,v,+} + E_{2q} - \hbar\omega_{l_1})(E_{k'+q,c,+} - E_{k'+q-l_1,v,+} + \hbar\omega_{l_1})} \tag{3.2}$$

(The schemes of all the scattering processes can be obtained by the permutation of the localization of the operators H_{s-d} and H_{e-r} at the vertices of the diagram as well as the consideration of the two possible spin orientations of the operators.)

We thus obtain 24 matrix elements corresponding to the Raman processes of light scattering of the two-particle elementary excitations of the type: the magnon-spin flip of the conduction electron. In our further considerations we will resort to the fact that the

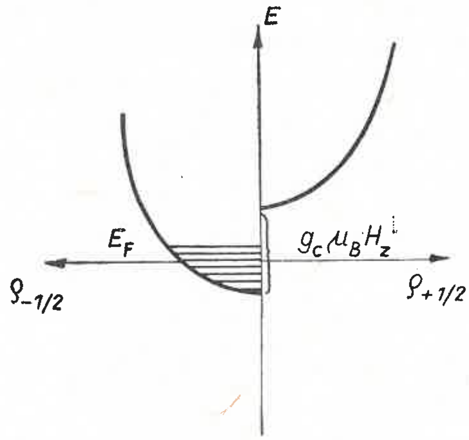

Fig. 1. Scheme of the energy levels of the conduction electrons in the external magnetic field. $\varrho_{-\frac{1}{2}}$ and $\varrho_{+\frac{1}{2}}$ denote the density of the conduction band states for the spin $s = -\frac{1}{2}$ and $+\frac{1}{2}$, respectively. H_z stand for the static homogeneous magnetic field, E_F denotes the Fermi level

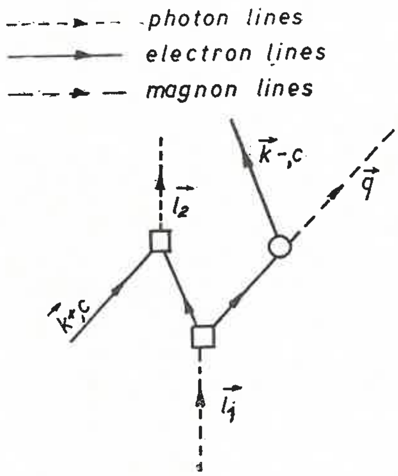

Fig. 2. Illustration of the scattering processes giving rise to the Raman absorption due to the magnon-spin flip of the conduction electron excitations

difference between the width of the energy gap E_g and the energy of incident photons is considerably larger than the s–d exchange integral in many experiments $E_g - \hbar\omega_{l_1} = E_{k,c} - E_{k,v} - \hbar\omega_{l_1}$ is of the order of 0.5 — 1 eV, whereas the exchange integral estimated due to the transport phenomena is of the order of 0.01–0.1 eV [14, 17]. The above simplifications allow for the representation of the $W(t)$ for the Raman light scattering of the magnon with the simultaneous spin flip conduction electron, in the following way

$$W(t) = \frac{B}{\omega_{l_1}} \sum_{q,l_2,k} \left| \frac{u_{l_2} p_{k-q,v;k-q,c} u_{l_1} p_{k-q,c;k-q,v} J_{s-d}(\boldsymbol{k}-\boldsymbol{q}, \boldsymbol{k})}{(E_q - \hbar\omega_{l_2})(E_{2q} + E_{k-q,c,-} - E_{k,c,+})} \right|^2$$

$$\times \delta(\hbar\omega_{l_1} - \hbar\omega_{l_2} - E_{2q} + E_{k,c,+} - E_{k-q,c,-}) \tag{3.3}$$

where

$$B = \frac{8\pi^2 h e^4}{\varepsilon^2 V^2 m^4}. \tag{3.3a}$$

Due to E_q(3.3) we obtain differential extinction coefficient in the following form

$$\frac{d^2 h}{d\omega d\Omega} = \frac{e^4 h}{m^4 c^4 V} \sum_{q,k} \left[\frac{\overline{u_{l_2} p_{k-q,v;k-q,c} u_{l_1} p_{k-q,c;k-q,v}}}{E_q - \hbar\omega_{l_2}} \frac{J_{s-d}(\boldsymbol{k}-\boldsymbol{q}, \boldsymbol{k})}{E_{2q} + E_{k-q,c,-} - E_{k,c,+}} \right]^2$$

$$\times \delta(\hbar\omega_{l_1} - \hbar\omega_{l_2} - E_{2q} + E_{k,c,+} - E_{k-q,c,-}) \tag{3.4}$$

where the solid line over the product of the matrix elements means that we make use of the approximation of the weak dependence of this product on the wave vectors $\boldsymbol{l}_1$ and $\boldsymbol{l}_2$ which is usual in qualitative estimations [1, 7, 18, 19].

4. Conclusions

The analysis of Eqs (3.3) and (3.4) enables us to formulate the following conclusions:

a) In the antiferromagnetic semiconductor, due to the existence of two antiparallel sublattices of the localized spins, is the occurrence of the considered Raman effect which does not depend on the sign of the s–d exchange integral. Note that in the ferromagnetic semiconductors the magnon-spin flip processes are generated only in the case of the negative value of the s–d exchange integral i. e. for the antiparallel interaction between the spins of the localized and conduction electrons.

b) In the formula (3.4) the energy denominator in the expression

$$\frac{J_{s-d}(\boldsymbol{k}, \boldsymbol{k}-\boldsymbol{q})}{E_{2q} + E_{k-q,c,-} - E_{k,c,+}} = \frac{J_{s-d}(\boldsymbol{k}, \boldsymbol{k}-\boldsymbol{q})}{E_{2q} g_c \mu_B H_z + \frac{\hbar^2}{2m}(q^2 - 2\boldsymbol{k}\cdot\boldsymbol{q})} \quad \text{for } \boldsymbol{q} = \boldsymbol{0} \tag{4.1}$$

is smaller for two orders of value than in the ferromagnetic semiconductors [20] and the still more considerable contribution to the extinction coefficient is given by the magnons near the centre of the Brillouin zone. On the other hand, due to a smaller value of the energy denominator in (4.1), the extinction coefficient for the antiferromagnetic semiconductors can be about 10^2 times larger than that in the ferromagnets.

On assuming, similarly like other authors [12, 14, 19], that

$$E_q - \hbar\omega_{l_2} \approx 0.5-1.0\ \mathrm{eV}, \quad (p_{k,v;k,c}) \approx 3\cdot 10^{-20}\,\frac{\mathrm{g\cdot cm}}{\mathrm{s}}, \quad S = \tfrac{1}{2},$$

$$\frac{J_{s-d}(\boldsymbol{k}, \boldsymbol{k}-\boldsymbol{q})}{E_{2q}+E_{k,c,-}-E_{k,c,+}} \approx 10, \quad J_{s-d}(\boldsymbol{k}, \boldsymbol{k}-\boldsymbol{q}) \approx 0.01\ \mathrm{eV} \tag{4.2}$$

and taking into account that in the processes there take part magnons from approximately about 1/10 of the Brillouin zone, we obtain the following value of the extinction coefficient for the concentrations of the conduction electrons of the order of 10^{16} cm^{-3}

$$\frac{d^2h}{d\Omega d\omega} \approx 10^{-9}-10^{-10}\ \mathrm{cm}^{-1}\ \mathrm{sr}^{-1}. \tag{4.3}$$

c) The energy absorbed in the considered Raman effect is given from (3.3) in the following relations:

$$\hbar\omega = \hbar\omega_{l_1} - \hbar\omega_{l_2} \approx E_{2q}+E_{k,c,-}-E_{k,c,+} \tag{4.4}$$

which is of the order 10^{-4}–10^{-3} eV i. e. is considerably smaller that absorbed in the ferromagnetic semiconductors, and is equal to about 0.01–0.1 eV [20].

d) Analogously, like in the ferromagnetic semiconductors [20], the change of a position of the absorbtion maximum given by (4.4) in an external magnetic field allows us to determine $g_c - g_v$.

REFERENCES

[1] P. A. Fleury, R. Loudon, *Phys. Rev.* **166**, 514 (1968).
[2] N. M. Amer, Tai-chang Chiang, Y. R. Shen, *Phys. Rev. Lett.* **34**, 1454 (1975).
[3] R. W. Kinne, W. J. O'Sullivan, J. F. Ryan, J. F. Scott, *Phys. Rev.* **B11**, 1960 (1975).
[4] P. D. Loly, B. J. Choudhury, W. R. Fehlner, *Phys. Rev.* **B11**, 1996 (1975).
[5] Y. Yafet, *Phys. Rev.* **152**, 858 (1966).
[6] C. K. N. Patel, R. Slusher, *Phys. Rev.* **177**, 1200 (1969).
[7] P. M. Platzman, P. A. Wolff, *Waves and Interactions in Solid State Plasmas, Solid State Physics Suppl.* **13**, Academic Press 1973.
[8] R. L. Hollis, J. F. Ryan, J. F. Scott, *Phys. Rev. Lett.* **34**, 209 (1975).
[9] G. Harbeke, E. F. Steigmeier, Solid State Commun. **6**, 747 (1968).
[10] A. E. Meixner, R. E. Dietz, D. L. Rousseau, *Phys. Rev.* **7**, 3134 (1973).
[11] S. Tekippe, V. J. Silberstein, R. P. Dresselhaus, M. S. Aggarwal, *Prol. of the International Conference on the Application of High Magnetic Field in Semiconductors Physics*, Würtzburg, 24 July-2 August 1975.
[12] R. M. White, R. B. Woolsey, *Phys. Rev.* **176**, 908 (1968).
[13] T. Kasuya, *Proc. of 11 Inter. Conf. on the Phys. of Semiconductors*, PWN, Warszawa 1972.
[14] S. V. Vonsovsky, *Magnetizm*, Nauka, Moscow 1971.
[15] T. Kasuya, *Prog. Theor. Phys.* **A6**, 45 (1956).
[16] I. Sugihara, *J. Phys. Chem. Solids* **33**, 1365 (1972).
[17] S. Klama, *Acta Phys. Pol.* **A40**, 595 (1971) and references cited therein.
[18] P. M. Platzman, P. A. Wolff, N. Tzoar, *Phys. Rev.* **174**, 480 (1968).
[19] R. Loudon, *Proc. R. Soc.* **275**, 218 (1963).
[20] A. Stasch, *Acta Phys. Pol.* (to be published).