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### FREE CARRIER ABSORPTION OF RADIATION IN FERROMAGNETIC SEMICONDUCTORS\*

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<u>Résumé</u> - Nous avons étudié l'absorption des porteurs libres dans les semiconducteurs magnétiques. Pour satisfaire la condition de conservation de l'énergie et du moment nous avons considéré les mécanismes de diffusion suivants : phonons acoustiques, magnons et impuretés ionisées. Les résultats sont appliqués à l'interprétation des valeurs du coefficient d'absorption de EuO riche en Eu.

<u>Abstract</u> - We investigate the absorption of radiation by free carriers in ferromagnetic semiconductors. To allow energy-momentum conservation the following scattering mechanisms were considered : acoustical phonons, magnons and ionised impurities. The results are used to interpret absorption coefficient data of Eu-rich EuO.

In the last two decades the study of the physical properties of magnetic semiconductors has been the subject of much experimental and theoretical investigation. A most interesting aspect in these systems is the influence of magnetism on their optical and electrical properties, which makes their behavior rather peculiar when one compares with observations in non-magnetic semiconductors. As an example, Eu-rich samples of EuO show a striking insulator-metal transition /1/ as the temperature is decreased below the ferromagnetic-ordering temperature (T<sub>c</sub> around 69 K). It is evident that the appearance of magnetic ordering in the system is responsible for the delocalization of the electrons trapped at the oxygen vacancies.

The purpose of this paper is to investigate the absorption of radiation by free carriers which were injected into the conduction band during the insulator-metal transition. This problem has been the object of various experimental studies /2,3,7/ and, we believe, some theoretical effort is desirable. In one of these studies /3/, a classical theory (Drude model) has been used to derive the density and scattering time of the carriers as a function of temperature. However, some observed features can not be explained by the classical theory. In particular, for moderately or heavily doped samples the absorption coefficient increases more rapidly than  $\lambda^2$ , where  $\lambda$  is the wavelength of the radiation, as expected when

quantum effects are not negligible. As is well know /4/, if  $\hbar\omega > E_f$ , where  $\omega$  is the radiation frequency and E<sub>f</sub> is the Fermi energy, a quantum approach should be used to calculate the absorption coefficient  $\alpha$ . In this paper we use a quantum-kinetic approach to derive  $\alpha$ . To allow energy-momentum conservation the following scattering mechanisms were considered : acoustical phonons, magnons and ionised impurities. It is shown that the processes assisted by ionised impurities, together with a small phonon contribution, can account for the frequency and temperature dependences of  $\alpha$  /3/, in the frequency interval  $3\mu$  to  $10\mu_{*}$ The observed /5,7/ resonance. at higher frequencies, but below the band edge, can be explained by the processes assisted by magnons in which an electron spin flip occurs. This resonance seems to persist /6/ even when the electron is bound to an impurity center at temperatures above the insulatormetal transition.

The absorption coefficient is defined by  $\alpha = -\frac{n}{c} \frac{1}{N} \frac{dN}{dt}$ , where  $\eta$  is the refraction index of the crystal, c is the velocity of the light and N = n - n<sub>o</sub> is the departure of the number of photons from the thermal equilibrium value n<sub>o</sub> =  $(c^{\hbar\omega/kT}-1)^{-1}$ . The photon mode is characterized by wave vector  $\vec{q}_{o}$ , frequency  $\omega$ , polarization index  $\gamma = 1/2$ and polarization vector  $\vec{\epsilon}_{\gamma}$ . The time rate of \* Work partially supported by CNPq, FINEP and CAPES (Brasilian Agencies). change of the number of photons is described by the kinetic equation

$$-\frac{\mathrm{dn}}{\mathrm{dt}} = \sum_{\vec{k},\vec{k}'} \sum_{\sigma,\sigma'} \left[ P(\vec{k}\sigma,\vec{k}'\sigma') + P(\vec{k}\sigma,\vec{k}'\sigma') - P^{\dagger}(\vec{k}\sigma,\vec{k}'\sigma') - (1) \right] + P(\vec{k}\sigma,\vec{k}'\sigma') - (1)$$

$$- P^{\dagger}(\vec{k}\sigma,\vec{k}'\sigma') \times f(\vec{k}\sigma) \left[ 1 - f(\vec{k}'\sigma') \right],$$

where f ( $\vec{k}\sigma$ ) is the distribution function for electrons with wave vector  $\vec{k}$  and spin  $\sigma$  and the P's represent the probability amplitude for an electronic transition where the supers cript (subscript) indicates the emission (+) or absorption (-) of a photon (scattering excitation). The probability amplitudes are calculated using second-order perturbation theory.

In evaluating  $\alpha$  we have neglected the photon wave vector, i.e., the electron only gains or loses energy by interacting with the radiation field. On the other hand, by interacting with the scattering mechanism the electronomly gains or loses momentum, since the energies involved are much smaller than the photon energy. The carriers are considered to form a degenerate electron gas and the distribution functions are step functions. As a result of the s-f exchange interaction between the conduction electrons and the localized spins, the conduction band splits into a band of spin up (zero energy level) and another of spin down separated by  $J_{s-f}$ <S Z> ( in molecular field approximation /9/), where J is the s-f exchange coupling and  $\langle S^2 \rangle$  is the thermal average value of the localized spins (2 is the direction of the magnetization). If  $J_{s-f}$ <S<sup>2</sup>>><sup>E</sup>f, only electrons with spin up exist in the conduction band. This seems to be the situation of experimental interest /3/.We have also performed an average over all the possible polarization directions of the radiation. With these assumptions the following probability amplitudes were evaluated : i) Ionised impurity scattering: in this case we have assumed a screened coulomb potential in the Thomas-Fermi approximation /8/. The ionised impurity contribution for  $\alpha$  is found to be

$$\alpha_{\mathbf{I}} = \mathbf{A}_{\mathbf{i}} \left\{ \mathbf{E}_{\mathbf{f}} \ {}^{\underline{\ell}} \mathbf{n} \right. \left| \begin{array}{c} 2\mathbf{E}_{\mathbf{f}} + 2\sqrt{\mathbf{E}_{\mathbf{f}}} \left( \mathbf{E}_{\mathbf{f}} \stackrel{\underline{\hbar}\omega) + \underline{\hbar}\omega}{\mathbf{h}\omega} + \mathbf{E}_{\mathbf{0}} \right. \right| + \\ \left. + \left( \frac{\underline{\hbar}\omega}{2} + \mathbf{E}_{\mathbf{0}} \right) \left[ 2\mathbf{E}_{\mathbf{f}} + \sqrt{\mathbf{E}_{\mathbf{f}}} \left( \frac{\mathbf{E}_{\mathbf{f}} + \mathbf{h}\omega}{\mathbf{h}\omega} \right)^{2} \right] + 2\mathbf{n} \\ \left. + \left( \frac{\underline{\hbar}\omega}{2} + \mathbf{E}_{\mathbf{0}} \right) \left[ 2\mathbf{n} \right] \left[ \frac{\mathbf{E}_{\mathbf{f}} + \sqrt{\mathbf{E}_{\mathbf{f}}} \left( \frac{\mathbf{E}_{\mathbf{f}} + \mathbf{h}\omega}{\mathbf{h}\omega} \right)^{2} \right] + 2\mathbf{n} \\ \left. + 2\mathbf{n} \right] \left[ \frac{(\underline{\hbar}\omega + \mathbf{E}_{\mathbf{0}}) \sqrt{\mathbf{E}_{\mathbf{f}}} \left( \frac{\mathbf{E}_{\mathbf{f}} + \mathbf{h}\omega}{\mathbf{h}\omega} \right) + \left( \frac{\mathbf{\hbar}\omega - \mathbf{E}_{\mathbf{0}} \right) \mathbf{E}_{\mathbf{f}}}{(\underline{\hbar}\omega + \mathbf{E}_{\mathbf{0}}) \sqrt{\mathbf{E}_{\mathbf{f}}} \left( \frac{\mathbf{E}_{\mathbf{f}} + \mathbf{h}\omega}{\mathbf{h}\omega} \right) - \left( \frac{\mathbf{\hbar}\omega - \mathbf{E}_{\mathbf{0}} \right) \mathbf{E}_{\mathbf{f}}}{\mathbf{E}_{\mathbf{f}}} \right] \\ \left. - 2\sqrt{\mathbf{E}_{\mathbf{f}}} \left( \frac{\mathbf{E}_{\mathbf{f}} + \mathbf{h}\omega}{\mathbf{h}\omega} \right)^{2} \right\},$$
 (2)

where  $A_i = 64e_0^6/3c_0\epsilon^2\hbar^4\omega^3$ ,  $E_0 = \hbar^2\kappa^2/2m$ ,  $\kappa = (6\pi e^2M_0/\epsilon E_f)^{1/2}$  is the inverse of the screening radius, m and n<sub>0</sub> are the mass and density of the carriers respectively, and  $\epsilon$  is the dielectric constant (the other quantities have the usual meanings ). ii) Acoustical phonon scattering: we shall assume that the electron-phonon interaction is the deformation potential /8/. As  $\hbar\omega >$  $E_f$ , the minimum phonon wave vector participating in the scattering is  $(2m\omega/\hbar)^{1/2}$ , of order  $I0^7 cm^{-1}$ . Thus in calculating the phonon contribution for  $\alpha$  we have considered a constant phonon dispersion relation. The final result is

$$\alpha_{\rm P} = A_{\rm P} \left\{ \frac{(2E_{\rm f} + \hbar\omega)}{3} \left[ E_{\rm f} (E_{\rm f} + \hbar\omega) \right]^{3/2} + \frac{1}{3} \left[ (\hbar\omega)^2 \left( \frac{(2E_{\rm f} + \hbar\omega)}{8} \times \left[ E_{\rm f} (E_{\rm f} + \hbar\omega) \right]^{1/2} + \frac{(3)}{8} \right] \right]^{1/2} + \frac{(\hbar\omega)^2}{16} \left[ \frac{\hbar\omega}{2\sqrt{E_{\rm f}} (E_{\rm f} + \hbar\omega)^2 + 2E_{\rm f} + \hbar\omega} \right]^{1/2} + \frac{(\hbar\omega)^2}{16} \left[ \frac{\hbar\omega}{2\sqrt{E_{\rm f}} (E_{\rm f} + \hbar\omega)^2 + 2E_{\rm f} + \hbar\omega} \right]^{1/2} + \frac{(\hbar\omega)^2}{16} \left[ \frac{\hbar\omega}{2\sqrt{E_{\rm f}} (E_{\rm f} + \hbar\omega)^2 + 2E_{\rm f} + \hbar\omega} \right]^{1/2} + \frac{(\hbar\omega)^2}{16} \left[ \frac{\hbar\omega}{2\sqrt{E_{\rm f}} (E_{\rm f} + \hbar\omega)^2 + 2E_{\rm f} + \hbar\omega} \right]^{1/2} + \frac{(\hbar\omega)^2}{16} \left[ \frac{\hbar\omega}{2\sqrt{E_{\rm f}} (E_{\rm f} + \hbar\omega)^2 + 2E_{\rm f} + \hbar\omega} \right]^{1/2} + \frac{(\hbar\omega)^2}{16} \left[ \frac{\hbar\omega}{2\sqrt{E_{\rm f}} (E_{\rm f} + \hbar\omega)^2 + 2E_{\rm f} + \hbar\omega} \right]^{1/2} + \frac{(\hbar\omega)^2}{16} \left[ \frac{\hbar\omega}{2\sqrt{E_{\rm f}} (E_{\rm f} + \hbar\omega)^2 + 2E_{\rm f} + \hbar\omega} \right]^{1/2} + \frac{(\hbar\omega)^2}{16} \left[ \frac{(\hbar\omega)^2}{16} \left[ \frac{\hbar\omega}{2\sqrt{E_{\rm f}} (E_{\rm f} + \hbar\omega)^2 + 2E_{\rm f} + \hbar\omega} \right]^{1/2} + \frac{(\hbar\omega)^2}{16} \left[ \frac{(\hbar\omega)^2}{16} \left[ \frac{(\hbar\omega)^2}{16} \left[ \frac{(\hbar\omega)^2}{16} \left[ \frac{(\hbar\omega)^2}{16} \left[ \frac{(\mu\omega)^2}{16} \left[ \frac{(\mu\omega)^2}{1$$

where  $A_p = 8/(2N_{\dot{q}_0} + 1)m^3e^2\epsilon_1^2 / 3\pi^2 DC\eta\hbar^9\omega^3\omega\dot{q}_0$ ,  $\omega_{\dot{q}_0}^{\star}$  and  $N_{\dot{q}_0}^{\star}$  are the energy and number of phonons with wave vector  $\dot{q}_0$  ( $q_0$  is an effective wave vector satisfying  $(2m\omega/\hbar)^{1/2}$   $< q_0 <$  maximum of the Brillouin zone),  $\epsilon_1$ is the deformation potential and D is the crystal density.

iii) Magnon scattering: The electron system interacts with the localized spins through

the s-f exchange interaction /9/. By assuming a quadratic magnon dispersion relation  $W_q^+ = g\mu_B^+H_0^{/h} + 2ISa^2q^2$ , with  $k_B^T > W_q^+$ , we have

$$a_{M} = A_{M} \left\{ \left( 2E_{f} + A + X_{O} \right) \sqrt{E_{f}} \left( \frac{E_{f} + A}{F_{f}} - \frac{A^{2}}{2} \ln \left| \frac{2\sqrt{E_{f}(E_{f} + A)} + 2E_{f} + A}{A} \right| - \frac{2\sqrt{E_{f}(E_{f} + A)} + 2E_{f} + A}{A} \right|$$

$$- X_{O} E_{f} \ell_{n} \left| \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} \right| + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f}(E_{f} + A)} + B} + \frac{2E_{f} + 2\sqrt{E_{f}(E_{f} + A)} + B}{2E_{f} - 2\sqrt{E_{f} - 2\sqrt{E_{f} - 2}} + \frac{2}{2\sqrt{E_{f}(E_{f} + A)} + B} + \frac{2}{2\sqrt{E_{f} - 2\sqrt{E_{f} - 2}} + \frac{2}{2\sqrt{E_{f} - 2\sqrt{E_{f} - 2}} + \frac{2}{2\sqrt{E_{f} - 2\sqrt{E_{f} - 2}} + \frac{2}{2\sqrt{E_{f} - 2\sqrt{E_{f} - 2}}$$

$$-\frac{X_{O}B'(A'-B')}{\sqrt{B'(B'-A)}} l_{n}$$

$$\frac{(A-B')E_{f} - \sqrt{B'(B'-A)E_{f}(E_{f}+A)}}{(A-B')E_{f} + \sqrt{B'(B'-A)E_{f}(E_{f}+A)}}$$
(4)

where 
$$A_M = J_{s-f}^2 e^{2} \max_B T/121c\pi^2 \eta h^6 \omega^3$$
,  $A = h\omega - J_{s-f}S$ ,  $B = A + X_0$ ,  $A' = AB/2X_0$ ,  
 $B' = B^2/4X_0$ ,  $X_0 = g\mu_B H_0^2/4mISa^2$ ,

 $J_{s-f}$  is the s-f exchange coupling, a is the lattice constant, I and S are the exchange constant and magnitude of the localized spins, H<sub>o</sub> is the applied magnetic field and T is the temperature.

These results will be used to interpret the data /3,7/ for the absorption coefficient of Eu-rich EuO. Let us take the following typical values for the parameters involved : D = 8.25g/cm<sup>3</sup>,  $\varepsilon = 25$ , a = 5 14A°,  $\varepsilon_1 = 10$  eV,  $J_{g-f}S = 0.35$ eV, n = 2.5, S=7/2,I = 0,67 k<sub>B</sub>,  $\omega_{q_o} = 260$  cm<sup>-1</sup> and H<sub>o</sub> =  $10^4$ G.

In figure, 1 we present a fitting to the data of Oliver et al/3/. The main contribution comes from the ionized impurity scattering, the phonon contribution acting as a background. The derived densities increase with decreasing temperature, as an indication of the insulator-metal transition. Figure 2 shows the effect of each contribution in a different frequency scale As we see the magnon effect goes to zero when  $\hbar\omega < J_{s-f}S - E_f$ . This represents a threshold frequency for the magnon assisted

processes, which involve electron spin flip transitions.



Fig. 1 : Absorption coefficient as a function of frequency. The lines are the theoretical results from the ionised impurity and phonon scatterings. Experimental points are from ref. /3/. The derived densities are (o)  $n_0 = 2.45 \times 10^{19}$ , (.)  $n_0 = 1.95 \times 10^{19}$ , ( $\Delta$ )  $n_0 = 1.20 \times 10^{15}$ ; ()  $n_0 = 8.00 \times 10^{18}$ , ()  $n_0 = 5.30^{\circ} \times 10^{18}$  (cm<sup>-3</sup>).



Fig. 2 : Illustration of the three scattering contributions for the absorption coefficient. Magnon (---); acoustical phonon  $(\cdot\cdot\cdot)$ , ionised impurity  $(-\cdot-\cdot)$ ; total contribution  $(---\cdot)$ .

The maximum effect occurs around  $\hbar\omega \gtrsim J_{S-f}S$ , reflecting the fact that all electrons with spin up are able to contribute to the absorption of radiation. Therefore, although the electrons have becomeitinerant, the magnon assisted process is a resonance like effect /7/, similar to that found /6/ when the electrons are bound to an impurity center at temperatures above the insulatormetal transition.

Note added in proof.- We thank Dr Godart for pointed out recent published data on reflectivity spectra of Eu rich EuO near the absorption edge (Escorne,M., Mauger,A. Godart,C., and Achard,J.C., J.Physique 40 (1979), 315).Their interpretation is in accord with the results of this paper concerning the magnon assisted process.

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