COMBINED RESONANCE IN CRYSTALS IN INHOMOGENEOUS MAGNETIC FIELDS

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We consider a new type of combined resonance, which arises in inhomogeneous static magnetic fields. Spatial inhomogeneity of the magnetic field results in a mixing of the motions with respect to coordinate and spin degrees of freedom. This in turn leads to spin transitions induced by the electric component of the high-frequency electromagnetic field. We calculate the intensity of combined resonance due to macroscopic inhomogeneity of the magnetic field or to microinhomogeneity of the spontaneous field in ferro- and antiferromagnets. It is shown that in both cases combined resonance may be much more intense than paramagnetic resonance. The conductivity-electron Hamiltonian which contains a periodic electrostatic potential and a spontaneous magnetic microfield, is transformed as part of the analysis into the effective-mass Hamiltonian, which contains two different mean homogeneous magnetic fields. One of the fields is identical with the induction and acts on the orbital motion of the electron, and the other appears in the spin term of the Hamiltonian and is itself dependent on the electron Bloch functions.

INTRODUCTION

It was shown earlier [1, 2] that spin-orbit interaction can lead to combined resonance of the band carriers in semiconductors, i.e., to the excitation of spin transitions by the electric field of the electromagnetic wave in the presence of a homogeneous static external magnetic field. Azbel [3] considered an effect which in some sense is the opposite, namely, the magnetic field of the electromagnetic wave in a metal is essentially inhomogeneous in the space near the surface, owing to the skin effect, and induces conduction-electron spin transitions that combine with the change in the state of orbital motion.

In this paper we consider spin transitions induced in the carriers of semiconductors, by the electric field of the electromagnetic wave. The mixing of the motions over the coordinate and spin degrees of freedom, necessary for such transitions, will be due not to the spin orbit coupling (as in [1, 2]), but to the inhomogeneity of the static magnetic field. This inhomogeneity can, for example, exist in an external static field, or be due to molecular or colloidal magnetic impurities.

1. COMBINED RESONANCE IN A MACROSCOPICALLY INHOMOGENEOUS MAGNETIC FIELD

The induction of the external static magnetic field will be denoted by $B + b(r)$, where $B$ is homogeneous and $b(r)$ is small compared with $B$. The mixing of the motion over the coordinate and spin degrees of freedom is brought about by the term $\beta (\sigma \cdot b)$ of the Hamiltonian of the electron, where $\beta$—effective spin magnetic moment of the electron ($\beta = g\beta \hbar/2$, $\beta = $ Bohr magneton). Inasmuch as $b(r)$ is of interest to us only as the cause of the mixing, we take account of it only in the aforementioned term of the Hamiltonian, and regard it as a small perturbation.

In the first order of perturbation theory, the matrix element of an arbitrary operator $\hat{L}(r)$, which depends only on the spatial coordinates of the electron, is equal to

$$<n + | \hat{L}(r) | n'-> = \beta \sum_I \left[ \frac{\langle \Psi_n | \hat{L} | \Psi_{n'} \rangle \langle \Psi_{n'} | b_I | \Psi_{n} \rangle}{E_{n'} - E_n - 2\beta B} ight.$$  
$$+ \frac{\langle \Psi_n | b_I | \Psi_{n'} \rangle \langle \Psi_{n'} | | \hat{L} | \Psi_n \rangle}{E_{n'} - E_n + 2\beta B} \right], \tag{1}$$

where $\Psi_n$ and $E_n$—states and Landau levels for the coordinate part of the Hamiltonian (for $b = 0$), $n$—aggregate of the three quantum numbers,
b_2 = b_X - i b_Y, and ± denotes the spin states of the electrons with \( \sigma_Z = \pm 1 \). The \( \Omega_z \) axis is chosen parallel to \( B \). For simplicity the band is assumed to be nondegenerate and the effective mass \( m \) and the \( g \)-factor are assumed isotropic.

To determine the intensity of the combined resonance it is necessary to calculate with the aid of (1) \( \langle n' | [\mathbf{\gamma}, | n' \rangle \rangle \), where \( \mathbf{\gamma} = m^{-1} (-i \mathbf{\nabla} + e \mathbf{A}/c) \) — electron velocity operator, \( \mathbf{A} = (0, \mathbf{B} \times 0) \) — vector potential of the homogeneous field \( B \). In the case of arbitrary \( b_2(r) \), the electromagnetic-wave absorption spectrum is continuous, since arbitrary increments of the kinetic energy of the electron in the \( \Omega_z \) direction are possible in the quantum transition. However, the spectrum breaks up into narrow bands if \( b_2(r) \) varies as a function of \( z \) smoothly enough to make

\[
\frac{1}{\omega_c} \sqrt{\frac{W}{m}} \left| \frac{\partial b_2}{\partial z} \right| \ll 1 \text{ and } \gamma,
\]

or else has the same period as the lattice: here \( \omega_c = eB/mc \) — cyclotron frequency, \( \gamma = 2\beta B/n\omega_c \), and \( W \) is equal to \( kT \) in nondegenerate semiconductors and to the Fermi energy in degenerate semiconductors. The frequencies that can be observed in this case are \( \omega = \omega_c |p \pm \gamma| \), where \( p \) is an integer.

By way of an example let us consider the case when \( b_2(r) \) is a linear function of the coordinate. We assume (for \( n = n' \))

\[
\langle + | \mathbf{\gamma}| - \rangle = i \omega_c \langle + | \mathbf{\gamma}| - \rangle, \quad X_{1,2} = \frac{1}{2} (r \pm i y),
\]

\[
X_3 = z, \quad r_B = (eh/eB)^{1/2} \left( \approx 10^{-3} / \mathbf{B}^{1/2},
\right.

if \( B \) is in Gauss).

\[ \text{(2)} \]

Only the matrix elements of the pure spin transitions differ from zero (\( n \neq n' \), \( \omega = \omega_c |\gamma| \)); they are

\[
\langle + | X_{1,2} | - \rangle = \pm \frac{1}{4} \frac{1}{\omega} \frac{\partial b_2 / \partial X_{1,2}}{B / r_B},
\]

\[
\langle + | X_3 | - \rangle = \pm \frac{1}{2} \frac{1}{\omega} \frac{\partial b_2 / \partial X_3}{B / r_B}.
\]

\[ \text{(3)} \]

The functions \( \varphi_n \) were normalized to unity in the principal region of cyclicity. In calculating the quantities (3) by means of formula (1), special caution must be exercised in the analysis of the matrix elements in which the states \( n \) and \( f \) have identical oscillator quantum numbers. These terms, which contain improper integrals, are best eliminated by using the commutation relations between the components of \( \mathbf{\gamma} \) and \( \mathbf{r} \).

If \( \partial b_2 / \partial z = 0 \), then it follows from \( \text{curl} \; b = \text{div} \; b = 0 \) that \( \partial b_2 / \partial X_2 = 0 \), i.e., the absorption vanishes at one of the circular polarizations of the wave in the \( xy \) plane.

To estimate the intensity of the combined resonance in question, it is convenient to compare the intensity with that of ordinary paramagnetic resonance at equal amplitudes of the electric field intensity of the wave in the former case and of the magnetic field in the latter. The ratio of these intensities is equal to

\[
\eta_i = \left| \langle + | X_i | - \rangle \right|^2 \frac{g\lambda}{\lambda}
\]

where \( \lambda = 2\beta e/e \) is the Compton wavelength of the electron. For example, if \( |\nabla b_2| / B = 10 \text{ cm}^{-1} \) and \( r_B \approx 10^{-5} \), then we get \( \eta_1 \approx 10^2 - 10^3 \). The quantities \( \eta_1 \) and \( \eta_3 \) can be appreciably larger if \( \gamma \approx 1 \) and \( |\gamma| < 1 \), respectively. Thus, the combined resonance in question, which coincides in frequency with the paramagnetic resonance, can greatly exceed the latter in intensity.

2. COMBINED RESONANCE IN SPONTANEOUS MAGNETIC FIELD OF FERROMAGNETS AND ANTFERROMAGNETS

In this section we neglect completely the spin-orbit interaction, i.e., we ignore it both as a cause of the mixing of the motions of the band electron over the coordinate and spin degrees of freedom, and as a cause of splitting of the bands and the \( g \)-shift (\( g = 2 \)). We do, however, take into account the interaction of the electron spin with the magnetic field of the spin and orbital momenta of the other electrons. In ferro- and antiferromagnets the latter can be represented by simple models in the form of a spontaneous magnetic field oscillating with the same periods as the lattice. This field, together with the external statistical field, constitutes the total magnetic microfield \( h(r) \). It is assumed below that the conduction band is nondegenerate, but it can have a multivalley structure.

We start with the Pauli one-electron Hamiltonian, which includes the periodic electrostatic potential of the lattice and the magnetic field \( h(r) \), and then go over to the representation of the effective-mass method. This is done by a transformation in which the expansion of the electron wave function in terms of the Bloch waves goes over into an expansion in plane waves. We assume that all the inter-band elements in the energy matrix are small, since they contain either the quasi-momentum \( k \) or the magnetic field \( h \). These inter-band elements can be approximately eliminated in
the usual manner\[^7\] with the aid of a canonical transformation in both the coordinate and the spin parts of the Hamiltonian. As a result we obtain the Hamiltonian of the effective-mass method

\[
H = -\frac{\hbar^2}{2} \sum_{ij} (m^{-1})_{ij} \hat{h}_i \hat{h}_j + \beta_0 \sigma \mathcal{B}(k_0) + (\mathbf{kV}_k) \mathcal{B}(k_0),
\]

where \(m^{-1}\) is the tensor of the reciprocal effective mass, \(k = -i \nabla + e A/\hbar\), and \(\mathbf{A}\) vector potential of the macroscopic induction \(\mathbf{B} = \hat{h}\):

\[
\mathcal{B}(k_0 + k) = |u_{k_0 + k}(r)|^2 \hat{h}(r),
\]

\[
\mathbf{V}_k \mathcal{B} = |\mathbf{V}_k \mathcal{B}(k_0 + k)|_{k \to 0}.
\]

Here \(u_{k_0 + k}(r)\) — periodic multiplier of the Bloch function, normalized such that \(|u_{k_0 + k}|^2 = 1\). The superior bar denotes averaging over the unit-cell volume; \(k_0 + k\) — total quasimomentum of the electron, \(k_0\) — quasimomentum value corresponding to the minimum of the energy in the valley in question.

Let us dwell in somewhat greater detail on an analysis of (4). Usually the transition to the presentation of the effective-mass method makes it possible to simplify the solution of problems involving the motion of an electron in smooth external fields. The field \(\hat{h}(r)\) is not smooth; however, since it has the same period as the crystal lattice and since the magnetic energy \(\beta_0 \mathcal{B}\) is small compared with the distance between bands, this magnetic field leads to the appearance in the Hamiltonian (4) of new terms which have a simple purely macroscopic structure. The first of these, \(\beta_0 \mathcal{B}\), describes the effective spin energy in the magnetic field, and the second \(\beta_0 \mathcal{B} (\mathbf{k} \cdot \nabla k_0) \mathcal{B}\) — the mixing of the motions over the coordinate and spin degrees of freedom in a homogeneous magnet.

An unexpected singularity of the operator (4) is that \(\mathcal{B}\) and the field \(\mathbf{B}\) which enters in \(\mathbf{k}\) are different mean macroscopic fields. Namely, \(\mathbf{B}\) is obtained from \(\hat{h}(r)\) by means of the usual averaging, and is the macroscopic magnetic induction, whereas \(\mathcal{B}\) is defined by (5) and is consequently essentially dependent on the distribution of the electron density in the Bloch states of the band electron with quasimomentum close to \(k_0\). The fact that the orbital-motion Hamiltonian should contain the induction \(\mathbf{B}\) has already been experimentally verified\[^8\] and theoretically justified\[^9\]. The field \(\mathbf{B}\), naturally, does not depend on the states of the electrons, whereas \(\mathcal{B}\) is different for electrons of different valleys. Inasmuch as the symmetry with respect to time reversal implies that \(\nabla_k |u_{k_0 + k}|^2\) vanishes when \(k_0 + k = 0\), we find that \(\nabla_k \mathcal{B} = 0\) for the valley with \(k_0 = 0\), i.e., the last term of (4), which leads to mixing of the motions over the coordinate and spin degrees of freedom, vanishes. In the same case, \(k_0 = 0\), we have in a compensated antiferromagnet \(\mathcal{B} = \mathbf{B}\).

We shall consider below two cases of combined resonance in magnets.

I. Single-domain magnet with \(k_0 \neq 0\). In this case the mixing of the coordinate and spin motions is realized by the last term of the operator (4). We confine ourselves for simplicity to the case of an isotropic effective mass of the conduction electron (in reality, however, this is most frequently not the case). The velocity operator, the matrix elements of which determine the probabilities of the quantum transitions of the electron, is in this case equal to

\[
\hat{v} = \frac{1}{i\hbar} [\hat{r} \mathcal{H}] = \frac{\hbar}{m} \mathbf{k} + \frac{\beta_0}{\hbar} \mathbf{V}_k (\mathbf{A} \mathcal{B}).
\]

The matrix elements \(\hat{v}\) can be easily calculated by using a formula similar to (1), with \(\mathbf{L} = \mathbf{A} \mathcal{B}\), and by regarding the last term in the square bracket of (4) as a perturbation \(b = (\mathbf{k} \cdot \sigma) \mathcal{B}\). If we use relations (2), we obtain for pure spin transitions

\[
\langle + | X_{1,1} | - \rangle = \pm \frac{i}{2\hbar} \mathcal{B} \frac{\partial \mathcal{B}}{\partial k_0},
\]

\[
\langle + | X_{1,1} | + \rangle = \pm \frac{i}{2\hbar} \mathcal{B} \frac{\partial \mathcal{B}}{\partial k_0},
\]

\[
B_0 = B_{0z} - iB_{0y}, \quad K_{0 z} = \frac{1}{i}(k_{0z} \pm i k_{0y}), \quad K_{0 y} = k_{0y},
\]

\[
\Gamma = \frac{B_0 }{\hbar \omega_c} = \frac{e}{\hbar} \sqrt{2mcB}.
\]

In the derivation of these formulas it was assumed that the quantization axis of the orbital Landau motion is the vector \(\mathbf{B}\), along which is directed the Oz axis of the unprimed Cartesian frame; the quantization axis of the spin motion, on the other hand, is the vector \(\mathcal{B}\), along which is directed the Oz' axis of the primed system. The choice of the latter, consequently, is different for different valleys.

To compare the intensity of the combined resonance in question with the intensity of ordinary paramagnetic resonance we assume that in order of magnitude \(\mathcal{B} \approx d \mathcal{B}\) of the magnetic field and the magnetic field of the crystal, where \(d \ll 1\) is the lattice constant. Then \(\eta_1 \sim d^2 / L^2 \sim 10^6\).

II. Multidomain magnet with \(k_0 = 0\). If \(k_0 = 0\) then, as noted above, in a single-domain magnet there is no combined resonance in the approximation considered. However, such resonance is possible in multidomain magnets, since the mixing of the coordinate and the spin motions is realized as a result of the coordinate dependence of \(\mathcal{B}\) on going through the domain boundaries. Inasmuch as in a compensated antiferromagnet \(\mathcal{B} = \mathbf{B}\) when
k_0 = 0, and B is practically independent of the coordinates even in a single-domain crystal, no combined resonance should arise in it. We shall therefore consider below single-crystal ferromagnets and ferrimagnets.

Assume that the domains are plane-parallel layers separated by the planes x = const. We assume further that the external magnetic field is directed along the Oz axis, and the average spontaneous field in neighboring layers is directed alternately in the positive and negative Oy directions. In this case the spontaneous field makes no contribution to f_{3z}, i.e., f_{3z} = B_z. The perturbation b_2(r) which enters in formula (1) is now equal to f_{3y}(x) = -i f_{3y}(x). Within the limits of each domain, f_{3y} ± does not depend on x, but f_{3y} reverses sign on going through the domain boundary.

In order for the field f_{3y} to be regarded as a small perturbation of the Landau states, we assume that f_{3y} \ll B_z \approx B. We assume also that the thickness of the transition layer between the domains is much smaller than r_B defined by formula (2), making it possible to assume that the field f_{3y} experiences a jump on going through the boundary of the domain layers. Then we obtain for the pure spin transition of the electron at the lowest Landau level,

$$\langle + | X_4 | + \rangle = \frac{1}{2(2\pi)^{1/2}} \frac{r_B}{1 + \gamma} \left( \frac{r_B N}{L} \right)^{1/2},$$

$$\langle + | X_4 | - \rangle = 0.$$  

(9)

Here L—dimension of the crystal in the Ox direction and N—number of domain layers in it. The matrix elements (9) are already the mean squares over all possible positions of the center of the Landau oscillator in the volume of the crystal.

For |b/B| = 0.1, r_B = 0.5 \times 10^{-5} cm, r_B N/L = 0.01, and |1 ± \gamma| → 1, we obtain \eta_{1,2} = 7 \times 10^5.

In the case II, and also in the case considered in Sec. 1, the spatial homogeneity of the magnetic induction leads to a broadening of the resonance absorption band, owing to the difference between the transition frequencies at different points of the crystal. However, if the field b(r) is directed perpendicular to B, this undesirable effect is minimal, being not in first but in second order in b/B (if the g-factor is isotropic, or if B is directed along a principal axis of the g tensor).

The three cases considered above and the estimates made show that combined resonance due to the inhomogeneity of the magnetic field should possess a considerable intensity, which can greatly exceed the intensity of ordinary paramagnetic resonance. Therefore paramagnetic resonance may be masked by combined resonance.

The combined resonance due to the inhomogeneity of the static magnetic field can arise not only for band carriers but also for local electron centers.

References:


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