

The Theory of Stimulated Magnon Generation

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The generation of magnons in a ferroelectric containing impurities is investigated theoretically. Weakly coupled impurity spins are used as the active centers. Optical pumping is assumed. The threshold value of the pumping and the minimum impurity concentration necessary for generation are determined. The estimates indicate that the conditions required for generation can be achieved.

INTRODUCTION

THE development of laser physics has drawn attention to the possibility of generating elementary boson excitations in solids, resulting from the stimulated emission of these excitations. Thus, the indicated generation of phonons ("phaser") has been realized; the feasibility of exciton generation has been demonstrated.

The present article is devoted to a theoretical investigation of single-mode magnon generation. The case of a ferroelectric is investigated. Impurity spins are utilized as the active centers, i.e., impurity atoms for which the magnitude of the spin and the energy of the exchange interaction with the spins of the matrix differ from the corresponding quantities for the atoms of the matrix; in this connection the impurity spins are assumed to be weakly coupled, i.e., the exchange interaction between the impurity and the matrix is appreciably smaller than the exchange interaction of atoms in the matrix. The concentration of impurity centers is assumed to be small. Optical pumping of the active centers is considered.

1. DESCRIPTION OF THE SYSTEM

We consider the following energy-level scheme for the impurity atom, assuming for simplicity that the spin is equal to $1/2$ (see the accompanying figure). The arrows shown on the figure indicate the projection of the impurity spin on the direction of the spins of the matrix, which are directed upward in the present case. Levels 1 and 2 correspond to the ground state which is split with respect to spin, and levels 3 and 4 correspond to the first excited state, which is again split. The splitting of the levels is due to the effective magnetic field created by the ferroelectric's magnetization (it is assumed that the temperature T is considerably below the Curie temperature). The direction of the arrows corresponds to a positive exchange integral I' between the impurity and the matrix.

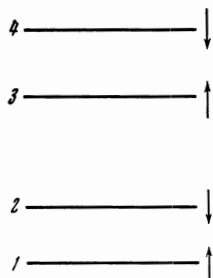


Diagram showing the energy levels of an active center.

The creation of a magnon can occur in the transitions $4 \rightarrow 3$, $2 \rightarrow 1$, and $4 \rightarrow 1$. Negative absorption can be obtained only in the transitions $4 \rightarrow 3$, $2 \rightarrow 1$; one should therefore regard these transitions as the operating ones. A small probability Γ_{ik} ($ik = 43, 21$) for the operating transition, which is proportional to the density of the magnon modes

$$\rho_M(\omega_{ik}) = \frac{1}{2} \omega_{ik} / \omega_M^{3/2}$$

is favorable in regard to obtaining negative absorption. Here $\omega_M = (6\pi^2)^{2/3} \cdot 2SI$ is the limiting magnon frequency, S is the spin of an atom in the matrix, and I is the exchange integral for the matrix atoms. Therefore, it is advisable to use the regime of low-frequency transitions: $\omega_{ik} \ll \omega_M$. This is realized in the case of a weakly coupled impurity spin.^[1] In the indicated case, upon taking Z nearest neighbors into account we obtain

$$\Gamma_{ik}^0 \cong \frac{\pi}{2S} (\omega_{ik})^2 \rho_M(\omega_{ik}), \quad \omega_{ik} = SZI'$$

Here and in what follows, Γ^0 denotes the spontaneous transition probability.

Optical pumping occurs in the transition $1 \rightarrow 4$. The transitions $4 \rightarrow 2$, $3 \rightarrow 1$ take place with the creation of phonons or photons. We shall assume that $\omega_{43}, \omega_{21} \ll \omega_{31}, \omega_{42}$. We emphasize that in general $\omega_{43} \neq \omega_{21}$ as a consequence of the different extent of overlap between the wavefunctions of the matrix atoms and the impurity atoms in the ground and excited states.

We shall consider the case when $T \ll \omega_{31}, \omega_{42}$.

2. THE CONDITIONS FOR GENERATION

The threshold condition for generation is the condition that the gain be equal to the losses:

$$c(n_i - n_k) b_{ik}^{jik} = \gamma^{jik}, \quad ik = 43, 21.$$

Here c is the concentration of active centers; n_i is the average occupation number of the i -th level;

$$b_{ik}^{jik} = \Gamma_{ik} / \rho_M(\omega_{ik}) \Delta_{ik}$$

denotes the probability for the spontaneous emission of a magnon in the j -th mode during the transition $i \rightarrow k$, Δ_{ik} is the half-width of the line; the magnon's attenuation is characterized by $\gamma^j = \gamma_0^j + c\gamma_1^j$, where the second term is due to the scattering of the magnon by the active centers. The number j_{ik} of the oscillating mode is determined from the condition that the ratio b_{ik}^{jik} / γ^j be a maximum.

The existence of the indicated maximum is necessary in order to obtain single-mode oscillation. The mode label j includes the magnon's frequency and the

other quantum numbers of the mode. Selection of the oscillating mode with respect to frequency is guaranteed by the line shape b_{ik} of the radiation; selection with respect to the remaining quantum numbers is guaranteed by the anisotropy of the magnon's attenuation γ (concerning the anisotropy of the attenuation, see for example^[2]). We note that the indicated anisotropy automatically imparts resonator-like properties to the medium.

The level widths are primarily determined by the transition involving these levels, i.e., by the longitudinal relaxation. Modulation broadening of these levels, i.e., transverse relaxation, is usually very much smaller.^[3] Therefore, for the system under consideration the pumping generally gives the important contribution to the level width. This property leads to a substantial complication of the theory.

The average occupation numbers are determined by the kinetic equations in the stationary regime. In this way we obtain the following result for the threshold value F of the photon flux associated with the pumping field:

$$F_{ik} = \frac{mc_p \Delta \omega}{4\pi^2 e^2 f D_{ik} A^1} (B_{ik} - D_{ik}(A^1 \Delta_{ik}^0 + A^0) - [B_{ik} - D_{ik}(A^1 \Delta_{ik}^0 + A^0)]^2 - 4D_{ik} A^1 (D_{ik} A^0 \Delta_{ik}^0 + C_{ik})^{1/2}), \quad ik = 43, 21. \quad (1)$$

Here e and m are the electron's charge and mass; c_p is the speed of light in the crystal at the pumping frequency; $\Delta \omega$ is the width of the spectral interval of the pumping field; f is the oscillator strength of the transition $1 \rightarrow 4$;

$$\begin{aligned} D_{ik} &= \gamma^{ik} \rho_M(\omega_{ik}) / c \Gamma_{ik}, & A^0 &= (\Gamma_{12} + \Gamma_{21})(\Gamma_3 \Gamma_1 - \Gamma_{31} \Gamma_{13}), \\ A^1 &= \Gamma_{13} \Gamma_{21} + \Gamma_3 (2\Gamma_{21} + \Gamma_{12} + \Gamma_{12}^0), & B_{13} &= \Gamma_{21} (\Gamma_{31}^0 - \Gamma_{13}^0), \\ & & B_{21} &= \Gamma_3 (\Gamma_{12}^0 - \Gamma_{21}^0), \\ C_{43} &= 0, & C_{21} &= \Gamma_{21}^0 (\Gamma_3 \Gamma_1 - \Gamma_{31} \Gamma_{13}), \\ \Delta_{13}^0 &= \Gamma_3 + \Gamma_4, & \Delta_{21}^0 &= \Gamma_{21} + \Gamma_{12}, & \Gamma_3 &= \Gamma_3 + \Gamma_{31}^0, \\ & & \Gamma_1 &= \Gamma_{13} + \Gamma_{12}^0 + \Gamma_{41}^0, \\ \Gamma_{ik} &= \Gamma_{ik}^0 (1 + \nu_{ik}); & \Gamma_{hi} &= \Gamma_{ik}^0 \nu_{ih}, \quad i > k, \\ \nu_{ik} &= [\exp(\omega_{ik} / T) - 1]^{-1}. \end{aligned}$$

The necessary condition for generation to be possible in the transition $i \rightarrow k$ is the condition that F_{ik} be real; this condition reduces to

$$F_{ik} > 0. \quad (2)$$

If this condition is satisfied for only one of the operating transitions, then generation will occur in that transition. However, if condition (2) is satisfied for both transitions, then generation starts in that transition which has the smallest value of F_{ik} .

The following necessary (but not sufficient) conditions for the transitions $4 \rightarrow 3$ and $2 \rightarrow 1$, respectively, follow from (2):

$$\Gamma_{34}^0 > \Gamma_{13}^0, \quad \Gamma_{12}^0 > \Gamma_{21}^0. \quad (3)$$

These are the conditions governing the feasibility of obtaining negative absorption in the corresponding transitions.

Upon fulfillment of (3), condition (2) reduces to

$$c > C_{ik} \equiv \gamma_{ik}^{ik} R_{ik} / (1 - \gamma_{ik}^{ik} R_{ik}), \quad (4)$$

where

$$\begin{aligned} R_{13} &= \rho_M(\omega_{13}) (\sqrt{A^1 \Delta_{13}^0} + \sqrt{A^0})^2 / B_{13} \Gamma_{13}, \\ R_{21} &= \rho_M(\omega_{21}) (A^1 \Delta_{21}^0 - A^0)^2 \Gamma_{21}^{-1} \{B_{21} (A^1 \Delta_{21}^0 + A^0) \\ &\quad + 2A^1 C_{21} - 2[A^1 (B_{21} \Delta_{21}^0 + C_{21}) (B_{21} A^0 + A^1 C_{21})]^{1/2}\}^{-1}. \end{aligned}$$

The necessary condition for generation in the transition $i \rightarrow k$, namely $\gamma_{ik}^{ik} R_{ik} < 1$, follows from Eq. (4). The quantity c_{ik} is the minimum concentration of active centers necessary for generation in the transition $i \rightarrow k$.

3. FAST TRANSITIONS $4 \rightarrow 2, 3 \rightarrow 1$

Let us consider the most timely case, when the transition rates $4 \rightarrow 2, 3 \rightarrow 1$ are large in comparison with the rates of the effective magnon transitions:

$$\Gamma_{42}^0, \Gamma_{31}^0 \gg \Gamma_{43}, \Gamma_{21}. \quad (5)$$

First let us consider the transition $4 \rightarrow 3$. From inequality (4) we obtain

$$\gamma_{13}^{43} \rho_M(\omega_{43}) \Gamma_{42}^0 (\Gamma_{42}^0 + \Gamma_{41}^0 + \Gamma_{31}^0) / \Gamma_{43} \Gamma_{21} < 4, \quad (6)$$

$$c > \gamma_{13}^{43} \rho_M(\omega_{43}) \Gamma_{42}^0 (\Gamma_{42}^0 + \Gamma_{41}^0 + \Gamma_{31}^0) / \Gamma_{43} \Gamma_{21}. \quad (7)$$

We shall assume that the quantity B_{ik} gives the major contribution to the radicand appearing in (1): This is favorable to a reduction of the threshold value of the pumping field. We obtain

$$F_{43} = \frac{mc_p \Delta \omega \gamma^{43} \rho_M(\omega_{43})}{4\pi^2 e^2 f} \frac{(\Gamma_{12} + \Gamma_{21})(\Gamma_{42}^0 + \Gamma_{41}^0)(\Gamma_{42}^0 + \Gamma_{41}^0 + \Gamma_{31}^0)}{\Gamma_{43} \Gamma_{21}} \quad (8)$$

The conditions for the validity of this formula are (6) and (7) in the sense of strong inequalities. It follows from (8) and from the indicated conditions that

$$F_{43} \ll \Gamma_{21}^0 (1 + 2\nu_{21}) \frac{\Gamma_{42}^0 + \Gamma_{41}^0}{\Gamma_{42}^0} \frac{mc_p \Delta \omega}{4\pi^2 e^2 f}$$

For the transition $2 \rightarrow 1$ we find

$$F_{21} = \frac{mc_p \Delta \omega}{4\pi^2 e^2 f} \Gamma_{21}^0 \frac{\Gamma_{42}^0 + \Gamma_{41}^0}{\Gamma_{42}^0}. \quad (9)$$

The conditions for the applicability of this formula are as follows:

$$2\gamma_{12}^{21} \rho_M(\omega_{21}) \ll 1, \quad c \gg 2\gamma_{12}^{21} \rho_M(\omega_{21}). \quad (10)$$

In deriving expressions (9) and (10) it was assumed that $\Gamma_{41}^0 \lesssim \Gamma_{42}^0$ and that ν_{21} is not too large.

We note that if the temperature is not too large in comparison with ω_{21} , then the pumping threshold for the transition $4 \rightarrow 3$ is considerably below that for the transition $2 \rightarrow 1$. However, conditions (6) and (7) are much more stringent than (10).

4. NUMERICAL ESTIMATES

We shall use the following values of the parameters in order to estimate the probabilities of single-magnon transitions: $s = 3, Z = 6, I = 10^{12} \text{ sec}^{-1}, I' = 5 \times 10^{10} \text{ sec}^{-1}$. Then $\omega_M = 10^{15} \text{ sec}^{-1}, \omega_{ik} = 10^{12} \text{ sec}^{-1}, \rho_M(\omega_{ik}) = 1.5 \times 10^{-15} \text{ sec}, \Gamma_{ik}^0 = 0.75 \times 10^9 \text{ sec}^{-1}, ik = 43, 21$. Condition (5) cannot be satisfied if the transitions $4 \rightarrow 2, 3 \rightarrow 1$ are photon transitions. This condition can be satisfied in the case of one-phonon transitions. In order to do this, the frequencies ω_{42} and ω_{31} must lie inside the phonon spectrum. Let us take $\Gamma_{31}^0 = \Gamma_{42}^0 = 10^{11} \text{ sec}^{-1}$. Finally, let $\gamma_{01k}^j = 10^{-2} \omega_{ik} = 10^{10} \text{ sec}^{-1}$. Condition (7) reduces to $c > 0.5$, so that generation in the transition $4 \rightarrow 3$ is impossible for the values of the parameters under consideration.

Let us assume that $\gamma_{12}^j = \sigma N \nu$, where σ is the cross section for the scattering of a magnon by an impurity

center, N is the number of cells per unit volume, and v is the magnon's velocity. Then inequality (10) reduces to $\sigma \ll 10^{-13} \text{ cm}^2$, $c \gg 10^{-5}$, which is certainly satisfied. Thus, generation is possible in the transition $2 \rightarrow 1$. Assuming $\Delta\omega = 10^{10} \text{ sec}^{-1}$ and $c_p = 10^{10} \text{ cm/sec}$, we obtain the following result for the threshold value of the pumping field: $F_{21} = 1.5 \times 10^{19} \text{ f}^{-1} \text{ sec}^{-1} \cdot \text{cm}^{-2}$. Assuming $f = 10^{-8}$ to 10^{-4} (an intercombination selection rule exists for the transition), we obtain $F_{21} = 1.5 \times (10^{23} \text{ to } 10^{27}) \text{ sec}^{-1} \cdot \text{cm}^{-2}$, which is attainable.

Yttrium iron garnet containing rare-earth impurities may serve as an example of the type of system considered in this work.

¹Yu. A. Izyumov and M. V. Medvedev, *Teoriya magnitoporyadochennykh kristallov s primesyami* (Theory of Magnetically-Ordered Crystals Containing Impurities), Nauka, 1970; Yu. Izyumov, *Proc. Phys. Soc. Lond.* **87**, 521 (1966).

²A. I. Akhiezer, V. G. Bar'yakhtar, and S. V. Peletminskii, *Spinovye volny* (Spin Waves), Nauka, 1967 [English Transl., North-Holland, 1968].

³M. A. Krivoglaz and V. F. Los', *Ukr. Fiz. Zh.* **15**, 85 (1970).