

NON LINEAR MAGNETOACOUSTIC EFFECTS

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ABSTRACT

Non-conducting magnets comprise the set of elastic and magnetic subsystems coupled with each other by magnetostrictive interaction, which, being, nonlinear, not only renormalizes a sound velocity but makes a sound-like mode strongly nonlinear. The effects are notably great (“gigantic”) in antiferromagnetics with a soft magnetic mode “effective” nonlinear moduli for ultrasound can exceed ones for usual solids by 10^3 - 10^4 . As a result, many acoustic analogues of the known nonlinear optics phenomena became practically attainable. It is concluded that antiferromagnet with a soft mode is an exclusively convenient object for experimental realization of various nonlinear dynamic effect including strongly nonlinear ones.

1. INTRODUCTION

Any dynamic system (if there is no dry friction in it) being under the action of time-variable external generalized force can be considered as linear in a first approximation, i.e. at a small amplitude of this forces: the system response is proportional to an amplitude of action. All real systems seem to be nonlinear in principle with the growth of external action this proportionality is violated sooner or later. It is this sooner or later that is the most significant in practice, in an experiment aspect. Indeed, any pendulum is a system with a noticeable nonlinearity at great angles of deviation from equilibrium position but for example, for a Foucault’s pendulum, though this nonlinearity might be demonstrated in principle.

Another example of “sufficiently linear” system is an arbitrary solid media: the sound velocity in the majority of crystals does not depend on a sound wave amplitude at practically attainable intensities of sound. Therefore, in order to realize nonlinear acoustic effects in solids at a limited external action value one should choose crystal with very high nonlinear elastic moduli- the most known example can be lithium (metha) niobate, LiNbO_3 and other ferroelectrics i.e crystals with spontaneous electric dipole moments.

The crystals with spontaneous magnetic moments, i.e Ferro, ferri- and antiferromagnets, turn out to be significant interest from the view-point of the nonlinear acoustics of solid media. As it has been found, the effects are specifically great (“gigantic”) in antiferromagnets with a soft magnetic mode.

2. THEORETICAL CONSIDERATIONS AND CALCULATIONS

Two subsystems “coexist” in magnetic non-conducting solids- the elastic one (its weak excitations are phonons) and the magnetic one (its excitations are describe by image of spin waves, i.e., magnons). When taking into account the magnetoelastic interactions between subsystems, weakly excited state of the system as a whole should be described by means of new normal oscillations (“quasi-particle”) mixed magnetoelastic waves. Here the natural problem arises under which conditions and to what extent these magnetoelastic waves are non-linear, i.e when and how one should bear in mind the dependence of their characteristics on their amplitude. The problem on nonlinearity of mixed oscillations of that or other nature is of rather common interest by itself. In order to demonstrate this, let us consider two subsystems of the magnetoelastic crystal –firstly, separately and then switch on the magnetoelastic coupling.

As it was shown by experience, the dynamics of the majority of elastic solids is linear in great extent (Zarembko L.K. et al 1970) in measure of the linearity of Hookes’ law, establishing relations between strain and mechanical stresses.

In general, this relation follows from expansion of this relation follows from expansion of the crystal elastic energy in series of components of strains tensor \hat{U} Landau L.D. et al 1965, Tucker J.W et al 1972, Dieulesaint E. et al 1974:

$$F_e = \hat{C}_2 \hat{U} \hat{U} + \hat{C}_3 \hat{U} \hat{U} \hat{U} + \dots + \hat{\sigma} \hat{U} \quad (1)$$

where

$$\hat{U} \equiv \frac{1}{2} \left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} + \frac{\partial U_l}{\partial x_i} \frac{\partial U_l}{\partial x_j} \right)$$

is the strain tensor which includes the “geometric” nonlinearity (it is described by the last term in \hat{U} and is very small): $2\hat{C}_2 = C_{ijkl}$, $6\hat{C}_3 = C_{ijklmn}$ are elastic modulus tensors of the second, third etc order (their components have 4, 6, etc indices, respectively) $\hat{\sigma}$ components is the tensor of external stresses. The condition for

use of expansion of equation (1) is obviously, the requirement that every successive addendum be considerably less than foregoing one. To simplify a qualitative analysis let us forget for a while the tensor character of variables and moduli.

$$F_e = C_2UU + C_3UUU + C_4UUUU + \dots \tag{2}$$

$$\equiv C_2UU(1 + \Gamma_1U + \Gamma_2UU + \dots)$$

where $\Gamma_1 = \frac{C_3}{C_2}$; $\Gamma_2 = \frac{C_4}{C_2}$; are nonlinearity coefficients of the first and second orders, respectively. It is

obvious that nonlinear effects are more expressive t, the higher the addenda Γ_1U and Γ_2U^2 are though, let us remind that the addenda should be: $\Gamma_2U \ll \Gamma_1U \ll 1$ for efficiency of expansion of equation (2). For usual solids $C_2 \sim 10^{12} \text{ cm}^3$, $C_3 \sim 10^{13} \text{ cm}^3$. $C_4 \sim 10^{14} \text{ cm}^3$ (Zarembó L.K. et al 1970), therefore, $\Gamma_1 \sim 10$ and $\Gamma_2 \sim 10^2$. But amplitude of a.c strains in solids being practically attainable in real ultra- and hyper sounds experiments are rather small: $U_a \sim 10^{-4}$ at the input ultrasound intensity $I_{in} = \rho v_s^3 U_a^2 \sim 10^2 \text{ W km}^{-2}$ that is hardly realized (v_s is sound velocity, ρ is the medium density). Thus the nonlinearity factors in the majority of solids and practically attainable amplitudes of a.c. strains produce the very small nonlinear terms $\Gamma_1U \sim 10^{-3}$ and $\Gamma_2U \sim 10^{-6}$ and thereby the observation of nonlinear acoustic effects is hampered (although the nonlinear acoustics of solids is being developed successfully (Dieulesaint E. et al, 1974), since the crystals exist in which Γ_1 and Γ_2 are abnormally high.

Single sublattice ferromagnet serve as a simple example for a spin-concentrated crystals. The behaviour of its spin (magnetic) subsystem is phenomenologically described by the Landau-lifshitz equation.

$$\dot{\vec{M}} = -\gamma [\vec{M} \vec{H}_{\vec{m}}], \quad H_{\vec{m}} = \frac{\delta F_{\vec{m}}}{\delta \vec{m}} \tag{3}$$

Where $F_{\vec{m}}$ is the magnetic energy density, and $\frac{\delta F_{\vec{m}}}{\delta \vec{m}}$ is its variational derivative. This equation is nonlinear in its

structure, since the effective field $\vec{H}_{\vec{m}}$ can depend on the variable of the problem, ie. On the magnetization \vec{m} . Let us evaluate the level of nonlinearity attainable for the simplest case: a thin ferromagnetic plate in a parallel magnetic field $\vec{H} // \vec{z}$. If x-axis is perpendicular to the plate, then the magnetization on deviation from the plate plane is accompanied with the growth of the magnetic energy, therefore

$$F_m = -(\vec{M} \vec{H}) + 2\pi M x^2 \tag{4}$$

i.e.

$$\vec{H}_{\vec{m}} = (-4\pi M_x, 0, H)$$

Bearing in mind that $|\vec{M}| = M_0$; $\vec{H} + 4\pi \vec{M}_0 = \vec{B}$ and for small precession angles

$$\vec{M} = (U_x, U_y, 1 - (U_x^2 + U_y^2)/2),$$

where U_x, U_y are directing cosines of the \vec{M} vector, we

obtain:

$$\dot{U}_x = -\gamma H U_y; \quad \dot{U}_y = \gamma B U_x \left[\left(1 - (U_x^2 + U_y^2) / 2 \right) \right] \tag{5}$$

hence at $H \sim 4\pi M_0 \sim \vec{B}$ and in the vicinity of the resonance frequency $\omega_0 = \gamma \sqrt{BH}$ it follows for

$U_x(t)$

$$\ddot{U}_x + \omega_0^2 U_x (1 + \Gamma_2 U_x^2) = 0$$

where $\Gamma_2 \sim 1$. High precession angles can be realized comparatively simply in non-conduction ferromagnets under resonance conditions $U_x \sim h / \Delta H \sim 10e / 10e$, where ΔH is the resonance line width and h is the amplitude of a.c magnetic field ($\vec{h} \perp \vec{H}$). As a result, comparatively great values of different types ($\Gamma_2 U^2 \sim 1, etc$) turn out to be experimentally achievable in magnets.

So, from the experimental view-point among two subsystems of a magnetic and the former possesses practically linear normal modes of oscillations (phonons), and the latter-strongly non-linear one (magnons). It is clear that the fact that new normal modes (mixed) will be nonlinear-both quasi magnons and quasi phonons and moreover, at the so called magneto acoustic resonance point, where the normal modes cannot be divided on quasi magnetic and quasi acoustic ones.

The value of this nonlinearity turns out to be estimated without detailed calculations, basing on linear experiments data only. For instance, one of resonance frequencies of antiferromagnet with anisotropy of an easy plane type depends on external magnetic in-plane field H and external stresses σ in the following way

$$\omega_{f_0} = \gamma \sqrt{H(H + H_D) + 2H_E H_S} \tag{6}$$

where H_s is the effective field of lattice deformations in electrons, and in this case

$$H_s = H_{s0} - R\sigma \tag{7}$$

where σ is the value of compression stress along H .

The formula below is valid for the velocity of long-wave ($K \ll a_0^{-1}$) quasi-sound in AFEP

$$V_{SH} = V_S \sqrt{1 - 2H_E H_{med} / (\omega_{fk} / \gamma)^2} \tag{8}$$

where H_{med} is the effective field of dynamic magnetoelastic interaction ($H_{med} \leq H_S$ depending on the quasi sound wavetype), $\omega_{fk}^2 = \omega_{0k}^2 + \omega_m^2 K^2$ and V_m is velocity of pure magnons.

From linear experiments it is known for example, for the hematite $2H_E \sim 2.10^7 Oe$; $H_S \sim H_{med} \sim 10e$ (Borovik et al 1964, Seavey et al 1972 Ozhogin V.I et al 1973), therefore, at $H \sim 1kO_e$ and $\sigma \rightarrow 0$ it follows

$$V_s^{-1} \frac{dV_s}{d\sigma} \cong -R \cdot 2H_E^2 H_{med} / \left(\frac{\omega_{f_0}}{\gamma} \right)^2 \left[\left(\frac{\omega_{f_0}}{\gamma} \right)^2 - 2H_E H_S \right] \cong -0.6(Kbar)^{-1} \tag{9}$$

From the other hand, the similar effect is also possible for a purely elastic crystal and is anharmonicity. Starting from equation (2) and using ratios for the sound velocity $V_S = (C_2 / \rho)^{1/2}$ and for the static deformation

$$U_C \cong \sigma / C_2, \text{ it can be estimated } V_S^{-1} \frac{dV_S}{d\sigma} \cong \frac{C_S}{2C_2^2} \sim 5.10^{-3} (Kbar)^{-1} \tag{10}$$

To develop a rigorous theory of anharmonism of sound-like magnetoelastic oscillations it is necessary to write a set of equations for the magnetic and elastic subsystems taking into account a binding magnetoelastic interaction.

$$\vec{M} = \vec{M}_1 + \vec{M} \text{ and } \vec{L} = \vec{M}_1 - \vec{M}_2$$

where as usually, \vec{M}_1 and \vec{M}_2 are sublattice magnetizations and one can assume often that their moduli are invariable.

$$| \vec{M}_1(\vec{\Gamma}, t) | = | \vec{M}_2(\vec{\Gamma}, t) | = M_0$$

Then, transforming the Landau-Lifshitz equation for \vec{M}_1 and \vec{M}_2 (Ozhogin et al 1970) and writing the equation of motion for the elastic displacement $\vec{U}(\vec{\Gamma}, t)$ of a unit cell centre of gravity in the continuous medium approximation, we obtain

$$-\gamma^{-1} \vec{M} = [\vec{M} \vec{H}_{\vec{m}}] + [\vec{L} \vec{H}_{\vec{L}}] + \vec{R}(m) \tag{11a}$$

$$-\gamma^{-1} \vec{L} = [\vec{M} \vec{H}_{\vec{L}}] + [\vec{L} \vec{H}_{\vec{m}}] + \vec{R}(L) \tag{11b}$$

$$\rho \ddot{\vec{U}} = \delta T_{ij} | \delta x_j + R_i^{(u)} \tag{11c}$$

with additional conditions

$$M^2 + L^2 = (2M_0)^2; (\vec{M} \vec{L}) = 0 \tag{12}$$

With the aid of which the number of independent variable is reduced to 7. Here, $\vec{H}m$ and $H \rightarrow$ are “effective field” for corresponding magnetic variables; T_{ij} is the tensor of mechanical stresses; $\vec{R}(M), R^{(L)}$ and $R^{(u)}$ are corresponding relaxation (viscous”) addenda.

If the energy density ρ is known as a function of variables of the problem, the external field H and external mechanical stresses δ_{ij} then, as usually, (Tucker J. W. et al 1972, Thurston R. W. et al 1964).

$$\vec{H}m = -\frac{\delta\rho}{\delta\vec{m}} + \frac{\delta}{\delta x_i} \frac{\delta\rho}{\delta(\delta\vec{m}/\delta x_i)} \tag{13a}$$

$$\vec{H}_L = -\frac{\delta\rho}{\delta\vec{L}} + \frac{\delta}{\delta x_i} \frac{\delta\rho}{\delta(\delta\vec{L}/\delta x_i)} \tag{13b}$$

$$T_{ij} = \delta\rho/\delta U_{ij} \tag{13c}$$

(the analysis of subtle details related to introduction of T_{ij} can be found in (Murnaghan F et al 1954). Relaxation addenda can be written in various ways, since at small damping (which is of the principal interest, in fact) the way of their accounting is not critical. For instance, the magnetic relaxation can be taken into account in the form (Ozhogin V. I. et al 1970):

$$\vec{R}^{(m)} = -\frac{\rho}{2M_0\gamma} [\vec{M}\dot{\vec{M}}] - \frac{\rho}{2M_0\gamma} [\vec{L}\dot{\vec{L}}] \tag{14a}$$

$$\vec{R}^{(L)} = -\frac{\rho}{2\gamma M_0} [\vec{L}\dot{\vec{M}}] - \frac{\rho}{2\gamma M_0} [\vec{M}\dot{\vec{L}}] \tag{14b}$$

and the viscous friction in the form (Tucker J. W et al 1972):

$$R_i^u = \delta T_{ij}/\delta x_j; T_{ij} = 2\eta_i \left(U_{ij} - \frac{1}{3} \delta_{ij} U_{kk} + \eta_2 U_{kk} \delta_{ij} \right) \tag{14c}$$

The sum total of ratios equation 11 divided by equation 14, with the given function $\rho(\vec{M}, \vec{L}, U_{ij}, \frac{\delta\vec{m}}{\delta x}, \frac{\delta\vec{L}}{\delta x}, H, \delta_{ij})$ formulates practically completely the problem on nonlinear processes in magnetoelastic antiferromagnetic crystals. Solutions of this problem, even in the case of infinite medium, are extremely complicated (and are rich in variety to an equal extent). Therefore, above all, we shall try to discover the class of substances where the nonlinear regime is attained readily that is already at low external actions.

Let us consider the two sublattice antiferromagnet with “easy plane” anisotropy, the normal to this plane being designated by a unit vector \vec{n} . The energy density in this instance can be written in the form:

$$\rho = 2M_0 \left\{ \frac{I}{2} Em^2 - D(M_x I_y - M_y I_x) + \frac{I}{2} A(\vec{n}\vec{I})^2 - \frac{E_w}{2} a_0^2 \left[\frac{\delta I_i}{\delta x_j} \right]^2 - \vec{m}H \right\} + \hat{C}^2 \hat{U}\hat{U} + I\hat{B}\vec{I}\hat{U} - \delta\hat{U} \tag{15}$$

Here $\vec{m} \equiv (\vec{M}_1 + \vec{M}_2)/2M_0, \vec{I} = (\vec{M}_1 - \vec{M}_2)/2M_0$ are ferro- and antiferromagnetic vectors (dimensionless) therefore the constants of interaction E, D, A, E_w are effective fields of exchange.

Now to assess quantitatively the value of nonlinear effects, let us realise the following programme Physica 86 – 88B, 979 (1977):

- (a) neglect the tensor character of values included in equation (15).
- (b) confine ourselves to consideration of only those components of magnetic vectors which belong to a low-frequency mode W_{fk} (a “quasi-ferromagnetic”, of f-mode (Ozhogin V. I. et al 1976), since only their coupling with even more low frequency sound oscillations can be not small;

(c) be interested only in the most low-frequency magnetoelastic oscillations, namely quasi-sound ones. For these frequencies ($\omega \ll \omega_{fk}$) one can assume $d/dt = 0$ in equation for magnetic variables and

alternating determinations $U^{(1)}$ can be written in terms of effective" elastic moduli:

$$P_{eff} = \hat{C}^{(2)} + \Delta\hat{C}_{eff}^{(3)}\hat{U}\hat{U}\hat{U} + \tag{16}$$

$$\text{where } \hat{C}_{eff}^{(2)} = \hat{C}^{(2)} + \Delta\hat{C}^{(2)}; \hat{C}_{eff}^{(3)} = \hat{C}^{(3)} + \Delta\hat{C}^{(3)}$$

If the direction of the field H being parallel to the "easy plane is chosen as an axis X and \vec{n} as an axis Z_y , then, by minimizing equation (15), we shall obtain in equilibrium

$$\vec{m}_0 = (m_0, 0, 0); I = (0, I_0, 0),$$

where $m_0 \cong (H_0 + \bar{D})/E; I_0 \cong 1$ (at $T \ll T_N$). Introducing small deviation from this equilibrium position $\vec{m} = \vec{m} - \vec{m}_0; \lambda = 1 - I_0$, the magnetoelastic energy can schematically be presented in the form

$$\rho_{me} = \hat{B}\hat{I}\hat{U} - B_\alpha 1_0^2(U_0 + U_\alpha^{(1)}) + B_\beta I_0 \lambda x U_\beta^{(1)} + B_e \lambda x (U_x + U_0^{(1)}) \tag{17}$$

where $U^{(1)} \ll U_0$ are those or other alternating components of the strains tensor, and $U_0 \cong -B_\alpha / C^2$ is the equilibrium strain (spontaneous Magnetostriction). For variables of f-mode, we obtain;

$$\gamma^{-1} \mu_y = -(H + D) U_z; \gamma^{-1} \lambda_x = E \mu_z \tag{18}$$

$$\gamma^{-1} \mu_z = H \mu_y + [2B_0(U_0 + U_0^{(1)}) - E_w a_0^2 K^2] \lambda_x - B_\beta U_\beta^{(1)} \tag{19}$$

where $\tilde{B} \cong \frac{B}{2N_0}$ are magnetoelastic moduli in magnetic field units. Assuming $\omega \ll \omega_{fk}$ that left parts of

these equations equal to zero $\left(\frac{d}{dt} \cong 0\right)$, we express λx in terms of $U^{(1)}$:

$$\lambda_{xk} \cong -U_\beta^{(1)} E \tilde{B}_\beta / [w_{fk}(\vec{\gamma}, t) / \gamma]^2 \cong -A_\lambda U_\beta^{(1)} - G_\lambda U_\beta^{(1)} U_j^{(1)} \tag{19}$$

$$A_\lambda \cong E \tilde{B}_\beta / (w_{fk} / \gamma)^4; G_\lambda = 2E^2 B_\beta \tilde{B}_\beta / (w_{fk} / \gamma)^4$$

Substituting (19) into (17) we obtain renormalization of the linear elastic moduli

$$\Delta C_{AF}^{(2)} \cong EB^2 / 2M_0 (w_{fk} / \gamma)^2 \tag{20a}$$

and nonlinear, as well

$$\Delta C_{AF}^{(3)} \cong 2B_\delta A_\lambda^2 \cong B_\beta G_\lambda \sim 2E^2 B^3 (2M_0)^2 (w_{fk} / \gamma)^4 \tag{20b}$$

Similar estimations performed for a ferromagnet do not contain exchange enhancement factors $\gamma E / w_{fk} \gg \rho$:

$$\Delta C_F^{(2)} \cong B^2 / M_0 (w_k / \gamma) \tag{21a}$$

$$\Delta C_F^{(3)} \cong 2B^3 / M_0^2 (w_k / \gamma)^2 \tag{21b}$$

The most convenient objects for the mentioned effects observation is hematite ($\alpha - Fe_2O_3$) - an opaque dielectric rhombohedral antiferromagnet (space group D_{sd}^6) $FeBO_3, T_N = 348K$, Isomorphic to hematite, being transparent in a visible light can be of keen interest likewise. Therefore, it is reasonable to retrace main stages of calculation of nonlinear magnetoacoustic effects just for rhombohedral antiferromagnets, but their awkwardness should not frighten, since the qualitative side of the problem has been discussed above. For these crystals, the last but one addenda in equation (15), are concretized by following

$$P_e = \hat{C}\hat{U}\hat{U} = \frac{1}{2}C_{11}(U_{xx}^2 - U_{yy}^2) + \frac{1}{2}C_{33}U_{zz}^2 + C_{12}U_{13}U_{xx}U_{yy} \quad (22a)$$

$$+ C_{13}(U_{xx} + U_{yy})U_{zz} + (C_{11} - C_{12})U_{xy}^2 + 2C_{44}(U_{xz}^2 + U_{yz}^2)$$

$$+ 2C_{14}[(U_{xx} - U_{yy})U_{yz} + 2U_{xy}U_{xz}]$$

$$\rho_{me} = \vec{I}\vec{B}\vec{I}\hat{U} = B_{11}(I_x^2U_{xx} + I_y^2U_{yy}) + B_{12}(I_x^2U_{yy} + I_y^2U_{xx}) +$$

$$2(B_{11} - B_{12})I_xI_yU_{xy} + B_{33}I_z^2U_{zz} + 2B_{44}(I_yI_zU_{yz} + I_xI_zU_{xz}) +$$

$$2B_{14}[(I_x^2 - I_y^2)U_{yz} + 2I_xI_yU_{xz}] + 2B_{41}[I_yI_z(U_{xx} - U_{yy}) + 2I_xU_{xy}] \quad (22b)$$

The z-axis was directed along the triple axis C_3 and the x-axis along the two fold axis U_2 of crystal.

3. RESULTS AND DISCUSSION

The calculation shows that in rhombohedral $AFEP$ to which hematite ($\alpha - Fe_2O_3$) belongs the ME interaction renormalizes nine third-order moduli, of which only two prove to be independent (in the case when the field \vec{H} is parallel to the binary axis $IIIU_2II_x$ (Ozhogin V. I. et al Zh.EKSP. Theor. Fiz, 73, 988 (1977) [Sov. Phys. JEIP 46(3), 523 (1977)]).

The moduli $C_{155}(H)$ and $C_{455}(H)$ can be chosen as independent. Their values and field dependence were determined for hematite in the “linear” experiment (Berezhnov V. V. et al. Fizika. Tverdogo Tela, 24, 46 1870 (1982); on change ΔV_t . In the velocity of the transverse sound with polarization U_2 and wave vector KC_3 with the crystal statistically deformed. For determination of any of the above moduli at the given value of H , mechanical stresses of at least two directions should be applied for example shifting σ_{yz} . The measurement results were treated by means of the following relations.

$$C_{155} \equiv 2[C_{44}(C_{11} - C_{12})R_{11}(H) + C_{14}C_{44}R_{\perp}(H)]$$

$$C_{455} \equiv 4C_{44}[C_{44}R_{\perp}(H) + 2C_{14}R_{11}(H)]$$

where $R_{11} \equiv -(\Delta C_{44}/C_{44})_{11}\sigma_{yz}$, $R_{\perp} \equiv (\Delta C_{44}/C_{44})_{\perp}\sigma_{yz}$,

$$\Delta C_{44} \equiv 2\rho V_t(H)\Delta V_t(H)$$

The authors of the paper (Berezhnov V. V. et al. Fizika Tverdogo Tela 24, N6, 1870 (1982) cited here have used the phase method for detecting the variations in the sound velocity. This method is insensible to changes in the effective damping which grows with increase in mixing of magnetic and elastic modes, that is for hematite –with decrease of the magnetic field.

4. CONCLUSION

The above consideration permits to suggest that in the case when the initial disturbance is not a solitary acoustic wave but a periodical one sound self-focusing may occur in the crystal. This effect is the acoustic analogue of light self-focusing in nonlinear optics. It was calculated in (Ozhogin V. I. et al 1989).

In nowadays fundamental research, a great interest has arisen in three dimensional nonlinear waves. The special properties of easy plane antiferromagnetic described in this article might make them suitable experimental objects for the solution of general problems of nonlinear wave physics

5. REFERENCES

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