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Translated by J. G. Adashko

Effect of fluctuations in the critical range in a second-order phase transition on a nonequilibrium phase transition

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(Submitted 11 December 1979)
Zh. Eksp. Teor. Fiz. **78**, 2384–2391 (June 1980)

The effect of fluctuations in the critical range, during phase transitions in ferroelectric materials and liquid crystals, on a nonequilibrium phase transition is considered in concrete cases, generation of light at impurity atoms and molecules. Two different mechanisms for this effect are considered. The first determines the effect of fluctuations of the phonon subsystem in ferroelectrics in the critical range on the threshold for generation at impurity atoms (ions). It is shown that, depending on the detuning of the generation frequency from the frequency of transition between the operating levels of the impurity atoms, the generation threshold may either increase (small detuning) or decrease (large detuning). It is the first case that has practical significance. The second mechanism determines the effect of fluctuations of orientation of the molecules of nematic liquid crystals, in the critical range, on the threshold for generation at impurity molecues and on the wavelength of the generated radiation. It is shown that the generation threshold drops on approach to the critical point. The wavelength also decreases. The pertinent temperature dependencies are found. The results are in qualitative agreement with experimental data [S. Kuroda and K. Kubota, Appl. Phys. Lett. **29**, 737 (1976); S. A. Akopyan, G. A. Bardanyan, G. A. Lyakhov, and Yu. S. Chilingaryan, Sov. Tech. Phys. Lett. **5**, 217 (1979)].

PACS numbers: 64.70.Kb, 64.70.Ew, 77.80. – e, 61.30. – v

1. INTRODUCTION

Substances are known in which there are second-order phase transitions kind and which, at the same time, can be used as operating media, for example in lasers. For such substances it is of interest to investigate the mutual influence of equilibrium and nonequilibrium phase transitions. An example is a transition through a generation threshold.

Examples of such substances are laser ferroelectrics, in which the generation occurs at the impurity-ion levels,[1,2] and also solutions of dye molecules in liquid crystals.[3,4] Experimental investigations are known in which the effect of fluctuations on the generation process was observed in the critical range during a phase transition in nematic liquid crystals. Thus a dependence of the generation threshold on the degree of proximity to the phase-transition point has been observed.[3,4] It was observed that on approach to the critical point (either from above or from below) the generation threshold decreased. A decrease of the wavelength of the generated radiation was also observed.

The present paper considers two possible mechanisms for the effect of fluctuations in the critical range on the process of generation of optical radiation. The first mechanism may manifest itself in ferroelectrics. In this case, the generation parameters change on ap-

 0038-5646/80/061199-05$02.40

proach to the critical point because of growing fluctuations of the phonon subsystem. This sort of mechanism was considered in a paper of Emel'yanov and the author.[5] The calculations made in that paper were based on application of perturbation theory and therefore are correct only at sufficiently large detuning $|\omega - \omega_{ab}|$ (ω_{ab} is the frequency of the operating transition, ω the generation frequency). It was shown that at sufficiently large detuning (see Section 2), the generation threshold decreases on approach to the critical point. Here results are presented that were obtained without use of perturbation theory. It follows from them that on approach to the critical point, the generation threshold may either increase (the small-detuning range) or decrease (at large detuning).

The second mechanism may manifest itself in solutions of molecules in liquid crystals. The effect of a phase transition in nematic liquid crystals on the generation threshold is due in this case to fluctuations of the tensor that describes the orientational ordering in liquid crystals. It is shown that this effect leads to a decrease of the generation threshold and also to a decrease of the wavelength of the generated radiation.

2. EFFECT OF A PHASE TRANSITION IN FERROELECTRICS ON THE GENERATION OF OPTICAL RADIATION AT IMPURITIES

We shall consider the process of generation at operating levels a, b of the impurity atoms (or ions) in ferroelectrics, with allowance for the influence of the phonon subsystem.

In order to calculate the polarization at the operating levels a, b, we must use the system of equations for the elements of the density matrix $f_{ab}, f_{ba}, D = f_a - f_b$:

$$\frac{\partial D}{\partial t} = \frac{2i}{\hbar}(\mathbf{d}_{ab}f_{ba} - f_{ab}\mathbf{d}_{ba})\mathbf{E}(\mathbf{R}, t) - \gamma(D - D^0), \tag{2.1}$$

$$\left(\frac{\partial}{\partial t} + \gamma_{ab} + i\omega_{ab}\right) f_{ab} = -\frac{i}{\hbar}\mathbf{d}_{ab}D\mathbf{E}(\mathbf{R}, t); \quad f_{ba} = f_{ab}^*. \tag{2.2}$$

Following Ref. 5, we supplement equation (2.2) for the function f_{ab} with a term that takes account of resonance interaction with the phonons:

$$\left(\frac{\partial}{\partial t} + \gamma_{ab} + i\omega_{ab}\right) f_{ab} = -\frac{i}{\hbar}\mathbf{d}_{ab}D\mathbf{E} - \frac{i}{\hbar}\mathbf{C}\mathbf{U}f_{ab}. \tag{2.3}$$

The vector quantity C describes the interaction of the impurity atoms with the phonon subsystem; U is the vector displacement of the lattice.

Besides the interaction taken into account in (2.3), one of the form $i\mathbf{C}\cdot\mathbf{U}D/\hbar$ is also possible. It does not give a resonance contribution at optical frequencies and therefore is disregarded here. This contribution is important, however, in the so-called vibron theory of phase transitions in ferroelectrics.[6,7]

We shall consider the effect on the process of generation of optical phonons. We shall denote by ω_i the characteristic frequency of oscillations of the ions of the lattice, and by $\tilde{\omega}_i$ the frequency of the soft mode (for $T > T_c$). Under the condition $\hbar\tilde{\omega}_i \ll kT$, the first two moments of the random function U are determined by the expressions

$$\langle \mathbf{U} \rangle = 0, \quad \langle (\delta \mathbf{U}(\mathbf{R}, t))^2 \rangle = 3kT/M\tilde{\omega}_i^2. \tag{2.4}$$

The equation for the fluctuation $\delta\mathbf{U}$ can be written in the form of a Langevin equation

$$d^2\delta\mathbf{U}/dt^2 + \Gamma d\delta\mathbf{U}/dt + \tilde{\omega}_i^2\delta\mathbf{U} = \mathbf{y}(\mathbf{R}, t). \tag{2.5}$$

At low frequencies ($\Omega \ll \Gamma$), the spectral density of the fluctuations $\delta\mathbf{U}$ is determined by the expression

$$(\delta\mathbf{U})_\Omega^2 = \frac{2\tilde{\omega}_i^2}{\Gamma}\frac{\langle (\delta\mathbf{U})^2 \rangle}{\Omega^2 + (\tilde{\omega}_i^2/\Gamma)^2}. \tag{2.6}$$

For the impurity atoms, when $\gamma_{ab} \sim 10^8 - 10^9$, the inequality

$$\tilde{\omega}_i^2/\Gamma \gg \gamma_{ab} \tag{2.7}$$

is satisfied. Under this condition, the spectrum of the fluctuations $\delta\mathbf{U}$ can be treated as white noise. Then with the aid of formula (2.6) we find the following expression for the time correlator of the fluctuations $\delta\mathbf{U}$:

$$\langle \delta\mathbf{U}\delta\mathbf{U} \rangle_\tau = 2\mathcal{D}\delta(\tau), \quad \mathcal{D} = \langle (\delta\mathbf{U})^2 \rangle / \tilde{\omega}_i^2/\Gamma. \tag{2.8}$$

In order to determine the effect of fluctuations of the phonon subsystem on the generation of laser radiation, we shall treat equation (2.3) as an equation for the random function f_{ab}. The random influence enters it parametrically. We shall carry out a calculation of the polarization, which is determined by the averaged function $\langle f_{ab} \rangle$. With the aid of (2.3) we find the following equation for it:

$$\left(\frac{\partial}{\partial t} + \gamma_{ab} + i\omega_{ab}\right) \langle f_{ab} \rangle = -\frac{i\mathbf{d}_{ab}}{\hbar} D^0 \langle \mathbf{E} \rangle - i\frac{\mathbf{C}}{\hbar} \langle \delta\mathbf{U}\delta f_{ab} \rangle. \tag{2.9}$$

We shall consider a state near the generation threshold; therefore on the right side of this equation, the function D is determined by a prescribed pumping $D^0 > 0$.

Equation (2.9) is not closed, since it contains the correlator of the fluctuations $\delta\mathbf{U}$ and δf_{ab}. The equation for δf_{ab} in the white-noise approximation [see the condition (2.7)] can be written in the form

$$\left(\frac{\partial}{\partial t} + i\omega_{ab}\right) \delta f_{ab} = -i\frac{\mathbf{C}}{\hbar}\delta\mathbf{U}\langle f_{ab}(t) \rangle,$$
$$\delta f_{ab}(t) = -i\frac{\mathbf{C}}{\hbar}\int_0^\infty \delta\mathbf{U}(t-\tau)\, d\tau \langle f_{ab}(t) \rangle. \tag{2.10}$$

Using the last expression in formula (2.8) for the time correlator of the fluctuations δU, we get the following expression for the second term in the right side of equation (2.9):

$$-i\frac{\mathbf{C}}{\hbar}\langle \delta\mathbf{U}\delta f_{ab} \rangle = -\frac{\mathbf{C}^2}{3\hbar^2}\mathcal{D}\langle f_{ab} \rangle. \tag{2.11}$$

It is valid in any approximation to the interaction of impurity atoms with the phonon subsystem. Using this result, we write a closed equation for the function $\langle f_{ab} \rangle$ (below, we shall omit the symbol $\langle \ldots \rangle$):

$$\left(\frac{\partial}{\partial t} + \bar{\gamma}_{ab} + i\omega_{ab}\right) f_{ab} = -i\frac{\mathbf{d}_{ab}}{\hbar} D^0 \mathbf{E}. \tag{2.12}$$

Here we have introduced the following notation for the effective damping:

$$\bar{\gamma}_{ab} = \gamma_{ab}\left(1 + \frac{C^2\mathcal{D}}{3\hbar^2\gamma_{ab}}\right) = \gamma_{ab}\left(1 + \frac{C^2\Gamma}{\hbar^2\gamma_{ab}}\frac{kT}{M\tilde{\omega}_i^4}\right). \tag{2.13}$$

We shall estimate the role of the term that is determined by the interaction with the phonon subsystem

(proportional to C^2). We use for this purpose the following numerical values of the parameters that occur in formula (2.13): $C=10^{-7}$ erg/cm, $\Gamma=10^{11}$ sec^{-1}, $\omega_i=3\times10^{13}$ sec^{-1}, $M=10^{-22}$ g, $\gamma_{ab}=10^9$ sec^{-1}, $T=10^2$ K. We then find that

$$\tilde{\gamma}_{ab}/\gamma_{ab}\approx(1+10^{-4}\omega_i^4/\tilde{\omega}_i^4). \tag{2.14}$$

Hence it follows that far from the critical point, the role of the interaction with phonons is not important. But already at $\tilde{\omega}_i=3\times10^{11}$ sec^{-1}, that is for insignificant "softening" of the mode, the second term in the right side is of order 10^4, and consequently the interaction with phonons is dominant.

To determine the effect of interaction of impurity atoms with the phonon subsystem at the generation threshold, we use the energy-balance equation

$$1/Q=|\operatorname{Im}\varepsilon|. \tag{2.15}$$

Here Q is the quality factor of the resonator, and the imaginary part of the permittivity at the operating transition $a-b$ is determined by the expression

$$\operatorname{Im}\varepsilon=-\frac{4\pi|d_{ab}|^2}{3\hbar}\frac{\tilde{\gamma}_{ab}}{(\omega-\omega_{ab})^2+\tilde{\gamma}_{ab}^2}nD^0,\quad D^0>0. \tag{2.16}$$

On substituting this expression in the energy-balance equation (2.15), we get the equation

$$\frac{1}{Q}=\frac{4\pi|d_{ab}|^2}{3\hbar}\frac{\tilde{\gamma}_{ab}}{(\omega-\omega_{ab})^2+\tilde{\gamma}_{ab}^2}nD^0, \tag{2.17}$$

from which can be found the threshold value of the expression nD^0, the product of the impurity-atom concentration and of the population difference with allowance for interaction with the phonon subsystem.

We see that the sign of the effect (raising or lowering of the generation threshold on approach to the phase-transition point) depends on the ratio of the detuning $|\omega-\omega_{ab}|$ and the linewidth γ_{ab}. Thus, for example, from zero detuning equation (2.17) takes the form

$$\frac{1}{Q}=\frac{4\pi|d_{ab}|^2}{3\hbar\tilde{\gamma}_{ab}}nD^0 \tag{2.18}$$

and consequently the threshold value of nD^0 rises on approach to the critical point according to the law

$$nD^0\propto\frac{\omega_i^4}{\tilde{\omega}_i^4}\propto\frac{1}{(T-T_c)^2}. \tag{2.19}$$

The reverse effect, that is decrease of the threshold value of nD^0, is possible only at large detuning, when $|\omega-\omega_{ab}|\gg\gamma_{ab}$. This situation, as a rule, is not energetically advantageous in lasers.

3. EFFECT OF A PHASE TRANSITION IN A LIQUID CRYSTAL ON THE THRESHOLD AND FREQUENCY OF GENERATION AT IMPURITY MOLECULES

The state of orientational ordering in nematic liquid crystals is described by the tensor

$$S_{ij}(\mathbf{R},t)=\int(\nu_i\nu_j-{}^1/_3\delta_{ij})f(\boldsymbol{\nu},\mathbf{R},t)\,d\boldsymbol{\nu}; \tag{3.1}$$

here $\boldsymbol{\nu}$ is a unit vector along the long axis of the molecule, and f is the corresponding distribution function. To describe a phase transition in a liquid crystal, one can use a tensor order parameter η_{ij}, which is defined as a quasiaverage value of the tensor S_{ij}. The usual average over an ensemble is zero both in the isotropic and in the crystalline phase; that is, $\langle S_{ij}\rangle=0$.

A phase transition can also be described, not by introducing an order parameter defined as a quasiaverage, but by discussing the transition on the basis of the correlator of random functions averaged over the volume of the system [cf. (2.15) in Ref. 8]: that is, on the basis of the behavior of the correlator

$$\langle S_{ij}(t)S_{kl}(t)\rangle;\quad S_{ij}(t)=\int S_{ij}(\mathbf{R},t)\frac{d\mathbf{R}}{V} \tag{3.2}$$

during a change of temperature.

For the random function S_{ij} one can write an appropriate Ginzburg-Landau equation. For temperatures below the critical, when the nonlinear term is not important, this equation has the form[9]

$$\frac{\partial S_{ij}}{\partial t}+aS_{ij}-g\frac{\partial^2 S_{ij}}{\partial\mathbf{R}^2}=y_{ij}(\mathbf{R},t);\quad a=a_0\frac{T-T_c}{T}. \tag{3.3}$$

The moments of the Langevin source are determined by the expressions

$$\langle y_{ij}\rangle=0;\quad \langle y_{ij}(\mathbf{R},t)y_{kl}(\mathbf{R}',t')\rangle=2\frac{\mathscr{D}}{n}I_{ijkl}\delta(\mathbf{R}-\mathbf{R}')\delta(t-t'). \tag{3.4}$$

Here we have introduced a symbol for the tensor

$$I_{ijkl}={}^1/_2[\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}-{}^2/_3\delta_{ij}\delta_{kl}], \tag{3.5}$$

$\mathscr{D}$ is the appropriate diffusion coefficient, characterizing the molecular motion in the liquid crystal.

With the aid of equation (3.3), we find expressions for the spectral densities of the components of the tensor S_{ij}:

$$(S_{ij}S_{kl})_{\omega,\mathbf{k}}=\frac{2\mathscr{D}I_{ijkl}}{\omega^2+(a+g\mathbf{k}^2)^2}, \tag{3.6}$$

$$(S_{ij}S_{kl})_{\mathbf{k}}=\frac{\mathscr{D}I_{ijkl}}{a+g\mathbf{k}^2}. \tag{3.7}$$

Below, we shall also require an expression for the correlator $\langle S_{ij}S_{kl}\rangle_\tau$. It can be obtained with the aid of formula (3.6).

In calculating the fluctuations of the field near the generation threshold, we may suppose that the width of the spectrum of the fluctuations S_{ij} is in significantly greater than the quantity $\omega_{ab}(|\operatorname{Im}\varepsilon|-1/Q)$. Under this condition, the time correlator is

$$\langle S_{ij}(\mathbf{R})S_{kl}(\mathbf{R})\rangle_\tau=(S_{ij}(\mathbf{R})S_{kl}(\mathbf{R}))_{\omega=0}\delta(\tau). \tag{3.8}$$

The expression for the noise intensity follows from formula (3.6):

$$(S_{ij}(\mathbf{R})S_{kl}(\mathbf{R}))_{\omega=0}=\frac{2\mathscr{D}I_{ijkl}}{(2\pi)^3}\int\frac{d\mathbf{k}}{(a+g\mathbf{k}^2)^2}. \tag{3.9}$$

To isolate the temperature dependence, we introduce the dimensionless variable of integration $x=k(g/a)^{1/2}$. It is then evident that on approach to the critical point, the integral increases as $1/\sqrt{a}$, and consequently the noise intensity

$$(S_{ij}(\mathbf{R})S_{kl}(\mathbf{R}))_{\omega=0}\propto(T-T_c)^{-1/2}. \tag{3.10}$$

To determine how fluctuations of the tensor $S_{ij}(\mathbf{R},t)$ affect the generation threshold, we use the equation for the amplitude of the laser radiation. At zero detuning it has the form

$$\frac{dE}{dt}+\frac{\omega_{ab}}{2}\left(\frac{1}{Q}+\operatorname{Im}\varepsilon_{tr}(\omega_{ab})\right)E=0. \tag{3.11}$$

Here $\mathrm{Im}\varepsilon_{tr}$ is the imaginary part of the permittivity of the impurity molecules at the frequency of the transition $a \to b$. At zero detuning, it is determined by the expression

$$\mathrm{Im}\,\varepsilon_{tr}(\omega_{ab}) = -\frac{4\pi e_i e_j (\mathbf{d}_{ba})_i (\mathbf{d}_{ab})_j}{\hbar\gamma_{ab}} nD^0; \tag{3.12}$$

here $\mathbf{e}$ is a unit vector along the direction of the field.

Without allowance for the effect of fluctuations of the orientational ordering tensor S_{ij}, the impurity system may be considered isotropic. Under this condition, the dipole-moment tensor has the form

$$(\mathbf{d}_{ba})_i (\mathbf{d}_{ab})_j = {}^1/_3 \delta_{ij} |\mathbf{d}_{ab}|^2. \tag{3.13}$$

With allowance for the orienting effect on the part of the solvent (liquid crystal), the structure of the dipole-moment tensor changes and takes the form

$$(\mathbf{d}_{ba})_i (\mathbf{d}_{ab})_j = {}^1/_3 \delta_{ij} |\mathbf{d}_{ab}|^2 + C_1 S_{ij}. \tag{3.14}$$

The constant C_1 describes the degree of orientational ordering of the molecules dissolved in the liquid crystal.

With use of the expression (3.14), the expression for the imaginary part of the permittivity (3.12) can be exhibited in the form

$$\mathrm{Im}\,\varepsilon_{tr}(\omega_{ab}) = \mathrm{Im}\,\varepsilon_{tr}^0 + \mathrm{Im}\,\delta\varepsilon_{tr}. \tag{3.15}$$

The second term, according to (3.14), is proportional to the tensor S_{ij}.

We substitute this expression in equation (3.11) and carry out the averaging. As a result we obtain the balance equation

$$1/Q = |\mathrm{Im}\,\varepsilon_{tr}^0| - \langle \mathrm{Im}\,\delta\varepsilon_{tr}\delta E\rangle / \langle E\rangle, \tag{3.16}$$

which is not closed.

The calculation of the correlator $\langle \mathrm{Im}\delta\varepsilon_{tr}\,\delta E\rangle$ is analogous to that carried out in the derivation of the expression (2.11). We give the result at once:

$$-\frac{\langle \mathrm{Im}\,\delta\varepsilon_{tr}\delta E\rangle}{\langle E\rangle} = \frac{\omega_{ab}}{4}\left(\frac{4\pi C_1 nD^0}{\hbar\gamma_{ab}}\right)^2 e_i e_j e_k e_l (S_{ij}(\mathbf{R})S_{kl}(\mathbf{R}))_{\omega=0}. \tag{3.17}$$

The energy-balance equation (3.16) can now be written in the form

$$\frac{1}{Q} = \frac{4\pi|\mathbf{d}_{ab}|^2}{3\hbar\gamma_{ab}} nD^0 + \frac{\omega_{ab}}{4}\left(\frac{4\pi C_1 nD^0}{\hbar\gamma_{ab}}\right)^2 e_i e_j e_k e_l (S_{ij}(\mathbf{R})S_{kl}(\mathbf{R}))_{\omega=0}. \tag{3.18}$$

Hence it follows that allowance for the orienting effect of the solvent leads to an effective increase of the square of the modulus of the matrix elements of the impurity molecules. As a consequence, the threshold value $(nD^0)_{tr}$ is lowered.

Far from the critical point [we use formula (3.10)], the lowering of the threshold on approach to the critical point proceeds according to the law

$$\Delta(nD^0)_{thr} \propto (T - T_c)^{-1/2}. \tag{3.19}$$

On further approach to the critical point, the rate of lowering of the generation threshold slows down; and in the region where the second term on the right side of equation (3.18) dominates,

$$(nD^0)_{thr} \propto (T - T_c)^{1/2}. \tag{3.20}$$

Thus we have explained how fluctuations of the tensor S_{ij} affect the generation threshold of laser radiation of impurity molecules in a liquid crystal. In experiments reported in Refs. 3 and 4, there was also observed a change (decrease) of the generation wavelength on approach to the phase-transition point.

For an explanation of this relation, we turn to the dispersion equation

$$(\omega_{ab}^2\,\mathrm{Re}\,\varepsilon(\omega_{ab}) - c^2\mathbf{k}^2)E = 0. \tag{3.21}$$

Here we are again considering the case of zero detuning.

Because of fluctuations of the orientation of the liquid-crystal molecules, the expression for the permittivity of the whole medium contains a fluctuational addition [cf. (3.15)]

$$\mathrm{Re}\,\varepsilon = \mathrm{Re}\,\varepsilon^0 + \mathrm{Re}\,\delta\varepsilon. \tag{3.22}$$

Thus the index of refraction in the dispersion equation is determined by the resultant polarizability of the impurity molecules and of the solvent. But at the frequency of transition for the impurities, the function $\mathrm{Re}\varepsilon_{tr} = 0$; therefore the principal role at small detuning is played by the polarization of the liquid-crystal solvent.

As is well known,[9-12] the permittivity tensor of a liquid crystal can be exhibited in the form

$$\varepsilon_{ij} = {}^1/_3(\varepsilon_\parallel + 2\varepsilon_\perp)\delta_{ij} + (\varepsilon_\parallel - \varepsilon_\perp)S_{ij}. \tag{3.23}$$

Here $\varepsilon_\parallel$ and $\varepsilon_\perp$ are the longitudinal and transverse (with respect to the direction of the director n) permittivities. Comparing formulas (3.22) and (3.23), we find

$$\mathrm{Re}\,\varepsilon^0 = {}^1/_3\mathrm{Re}(\varepsilon_\parallel + 2\varepsilon_\perp);\quad \mathrm{Re}\,\delta\varepsilon = \mathrm{Re}(\varepsilon_\parallel - \varepsilon_\perp)e_i e_j S_{ij}. \tag{3.24}$$

We average the dispersion equation (3.21). As a result, we again obtain an unclosed equation

$$\omega_{ab}^2(\mathrm{Re}\,\varepsilon^0 + \langle \mathrm{Re}\,\delta\varepsilon\delta E\rangle/\langle E\rangle) = c^2k^2. \tag{3.25}$$

The calculation of the correlator is carried out by the same method as above. We give the result:

$$\frac{\langle \mathrm{Re}\delta\varepsilon\delta E\rangle}{\langle E\rangle} = \frac{\omega_{ab}}{4}\mathrm{Re}(\varepsilon_\parallel - \varepsilon_\perp)\frac{4\pi C_1 nD^0}{\hbar\gamma_{ab}} e_i e_j e_k e_l (S_{ij}(\mathbf{R})S_{kl}(\mathbf{R}))_{\omega=0}. \tag{3.26}$$

We substitute this expression in equation (3.25) and exhibit the value of the wave vector in the form

$$k = \frac{\omega_0(\mathrm{Re}\,\varepsilon^0)^{1/2}}{c} + \Delta k = k_0 + \Delta k. \tag{3.27}$$

For the value of Δk, in the linear approximation, we find the expression

$$\Delta k = \frac{\omega_{ab}^3}{2c^2 k_0}\mathrm{Re}(\varepsilon_\parallel - \varepsilon_\perp)C_1\frac{\pi nD^0}{\hbar\gamma_{ab}} e_i e_j e_k e_l (S_{ij}(\mathbf{R})S_{kl}(\mathbf{R}))_{\omega=0}. \tag{3.28}$$

We see that the sign of the quantity Δk depends on the signs of two quantities: $\varepsilon_\parallel - \varepsilon_\perp$ and C_1. If the coupling constant C_1 has the same sign as the quantity $\varepsilon_\parallel - \varepsilon_\perp$ (it is this, of course, that is expected), then the quantity Δk is positive, and therefore the quantity $\Delta\lambda = -\Delta k/k_0^2$ is negative.

Thus on approach to the critical point, the wavelength of the laser radiation, provided $\mathrm{Re}(\varepsilon_\parallel - \varepsilon_\perp)C_1 > 0$, decreases. Far from the critical point, where in formula (3.28) the dependence of the threshold value on the temperature is not yet significant, we find that

$$-\Delta\lambda \propto (T - T_c)^{-1/2}. \tag{3.29}$$

With approach to the critical point, this dependence becomes weaker, and under the condition (3.20)

$$-\Delta\lambda \propto (T-T_c)^{1/4}. \qquad (3.30)$$

The two examples considered show how fluctuations in the critical range, during phase transitions of the second kind in ferroelectric materials and liquid crystals, affect the characteristics of laser radiation produced by impurity atoms and molecules. A deeper investigation of this problem is naturally of interest. Of particular interest is a study of the mutual influence of equilibrium and nonequilibrium phase transitions.

We take this occasion to thank G. A. Lyakov for discussion of the results of the research.

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Translated by W. F. Brown, Jr.

Large-amplitude spin waves and magnetic relaxation in the superfluid phases of ^{3}He

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(Submitted 13 December 1979)
Zh. Eksp. Teor. Fiz. **78**, 2392–2410 (June 1980)

Solutions in the form of large-amplitude spin waves are obtained in the long-wave limit for the spin-dynamics equations of superfluid ^{3}He. The dispersion laws of these waves in the A and B phases are obtained and their stability is investigated. The magnetic-relaxation diffusion mechanism due to the spatial nonuniformity of the magnetization distribution is considered. A method of measuring the spin-diffusion coefficient in the superfluid phases of ^{3}He is proposed.

PACS numbers: 67.50.Fi, 75.30. – m, 75.30.Ds, 75.30.Kz

1. INTRODUCTION

Experiments[1-3] show that the relaxation of the magnetization in superfluid ^{3}He to the equilibrium value from the initial nonequilibrium state can apparently not be attributed in all cases to the action of one mechanism. The experiments are presently interpreted usually by the only theoretically sufficiently well developed "intrinsic" mechanism of Legget and Takagi,[4] whose applicability is restricted by the requirement that the magnetization be uniformly distributed in space. This restriction is strong in those cases when the initial state is prepared by tipping the magnetization by a finite angle from the direction of the external magnetic field H_0. Even a small inhomogeneity of H_0, because of the difference between the Larmor frequencies at various points of space, leads after a sufficiently long time to a considerable nonuniformity in the distribution of the magnetization. Even more important is the fact that in the A phase the spatially uniform precession of the magnetization is unstable and a nonuniform distribution of the magnetization sets-in spontaneously even in an ideally uniform external field.[5] The onset of the nonuniformity, together with spin diffusion, leads to a rather effective mechanism of longitudinal relaxation in the superfluid ^{3}He.

A theoretical investigation of the spatially inhomogenous states calls in the general case for the solution of a rather complicated nonlinear system of partial differential equations (see, e.g., Ref. 6). The problem is simpler for weakly inhomogeneous states, where the influence of the spatial nonuniformity on the precession of the magnetization can be regarded as a small perturbation. This approach is applicable if the energy of the inhomogeneity of the condensate of the Cooper pairs is small compared with the magnetic energy $\sim\chi H_0^2$, where χ is the magnetic susceptibility of the ^{3}He. In connection with the investigation of the relaxation, we shall be interested in states in which the energy inhomogeneity is comparable in order of magnitude with the spin-orbit energy. For the customarily employed fields $H_0 \approx 200$–300 Oe, the condition of smallness of the spin-orbit energy is satisfied with good accuracy at all temperatures.

 0038-5646/80/061203-10$02.40