

terminated only by the conditions for the screening of the spontaneous induction, and does not depend on the filling of the conduction band.

Let us stop now to discuss the main consequences of the examined picture of the p-n junction.

1) A BaTiO<sub>3</sub> crystal connected in an electric circuit will behave like a p-n junction with symmetric current-voltage characteristic. The symmetry of the characteristic is a result of repolarization, which causes the current to flow in one direction relative to the p-n junction.

2) During repolarization, nuclei of the opposite phase grow through the crystal. Non-equilibrium free carriers, with density determined by (2), are concentrated on the ends of these nuclei, in regions on the order of  $l$ . At the instant when opposite ends of nuclei meet, recombination takes place and is accompanied by emission of light. The frequency of such emission can be of the order of the width of the forbidden band, corresponding to violet light in the case of BaTiO<sub>3</sub>. The emission should take place over the entire volume of the crystal and is flash-like. Work aimed at observing this emission is now under way.

3) Thin layers with anomalously high free-carrier density should exist near the surfaces of crystals not equipped with electrodes. Thus, the electric conductivity along the surface should be much higher than in the direction perpendicular to the surface.

The authors thank B. M. Vul, V. A. Rassushin, and N. A. Penin for a discussion of the results.

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#### EXCITATIONS OF EXCITON TYPE IN A GAS

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Submitted 3 October 1966

ZhETF Pis'ma 5, No. 1, 13-16, 1 January 1967

In this note we consider collective excitations in a gas, similar in character to Frenkel excitons in molecular crystals.

In a not too rarefied gas, when the emission wavelength at the transition frequency exceeds the distance between particles, the main role in the interaction between atoms is played by dipole-dipole interaction

$$V(r) = (d_1 d_2)/r^3 - 3(d_1 r)(d_2 r)/r^5, \quad (1)$$

and delay effects can frequently be neglected.

As will be shown below, there are two characteristic regions of atom-density values  $n$ . At low densities the dispersion of the oscillations is connected with thermal motion of the atoms, and the analysis can then be carried out in the self-consistent-field approximation.

At large  $n$ , the dispersion of the oscillations is connected with the finite distance between particles, and we are dealing with excitons in a disordered system. The state-density distribution function was obtained in this case by Vlasov and Fursov [1]. We also obtain below the characteristic frequency of the relaxation due to thermal motion of the atoms.

1. We consider first the case of small  $n$ . The equation of motion for the distribution function  $P(r, v, t)$  of the atomic dipoles is

$$i\left(\frac{\partial}{\partial t} + \gamma + \left(\mathbf{V} \frac{\partial}{\partial \mathbf{r}}\right) + i\omega_0\right) P = \frac{d^2}{\hbar} \mathbf{E} f(\mathbf{v}), \quad (2)$$

where  $\omega_0$  is the transition frequency,  $\gamma$  the relaxation frequency connected with the atomic collisions,  $d$  the matrix element of the dipole moment, and  $f(\mathbf{v})$  the atom velocity distribution function, which can be regarded as Maxwellian

$$f(\mathbf{v}) = (\sqrt{\pi} s)^{-3} e^{-v^2/s^2} \quad (3)$$

In the self-consistent-field approximation the electric field can be expressed in terms of the distribution function  $P$

$$\mathbf{E}(\mathbf{r}, t) = n \frac{\partial}{\partial \mathbf{r}} \int d^3 r' d^3 v (P(\mathbf{r}', \mathbf{v}, t) \frac{\partial}{\partial \mathbf{r}}) |\mathbf{r} - \mathbf{r}'|^{-1} + \text{c.c.} \quad (4)$$

From (2) and (4) we obtain for plane waves ( $\sim \exp i[\mathbf{k} \cdot \mathbf{r} - \omega t]$ ) the dispersion equation for the longitudinal and transverse oscillations:

$$\Omega_{\parallel, \perp} \int \frac{f(\mathbf{v}) d^3 v}{\omega_{\parallel, \perp} - \omega_0 - (\mathbf{k} \cdot \mathbf{v}) + i\gamma} = 1, \quad \Omega_{\parallel} = -\frac{2}{3} \Omega_0, \quad \Omega_{\perp} = \frac{1}{3} \Omega_0; \quad (5)$$

$$\Omega_0 = 4\pi n d^2 / \hbar.$$

From this we get approximately, for long waves ( $kD \ll 1$ ,  $D = s/\Omega$ ),

$$\begin{aligned} \text{Re } \omega &= \omega_0 + \Omega \left(1 + \frac{1}{2} k^2 D^2\right), \\ \text{Im } \omega &= -\gamma - \frac{\sqrt{\pi} \Omega}{kD} e^{-1/k^2 D^2} \end{aligned} \quad (6)$$

The relations for the longitudinal and transverse waves are obtained from (6) by replacing  $\Omega$  by  $\Omega_{\parallel}$  or  $\Omega_{\perp}$  respectively. The self-consistent-field approximation is valid if the radius of the "collective" forces  $D_0$  is large compared with the distance between particles:

$$n D_0^3 \gg 1. \quad (7)$$

It is interesting to note that condition (7) defines also the region of paired collisions of the atoms with excitation transfer, investigated in [2-7]. Indeed, the scattering amplitude  $r_0$ , determined from the condition  $V(r_0)t_0/\hbar \gtrsim 1$ , where  $t_0 = r_0/s$  is the collision time, is of the order of  $r_0 \sim d/\sqrt{s\hbar}$ . Thus, the paired-collision criterion  $nr_0^3 \ll 1$  coincides with inequality (7). This situation is analogous to the case of an ideal plasma when many particles are contained in a sphere of radius equal to the Debye radius.

2. At higher densities, when  $nr_0^3 \gg 1$ , the thermal motion of the atoms plays a secondary role. In this case one can speak of an exciton in a system of randomly arranged atoms. The dispersion of the excitons near the bottom of the band (small  $k$ ) is

$$\omega = \omega_0 + \Omega(1 + 0(k^2 n^{-2/3})). \quad (8)$$

To calculate accurately the coefficient of  $k^2$  it is necessary to take into account multiple scattering of excitations by atoms, a rather complicated problem. If, however, we confine ourselves to a simplified model, neglecting correlation in the magnitude and orientation of the dipoles, then the state-density distribution function can be obtained by a well known method [8]. To estimate the frequency of relaxation due to thermal motion of the atoms, we consider the distribution function  $W(\Delta, \dot{\Delta})$  for frequencies  $\Delta = \omega - \omega_0$  and "accelerations"  $\dot{\Delta} \equiv d\Delta/dt$ :

$$W(\Delta, \dot{\Delta}) = \frac{1}{(2\pi)^2} \int dt d\tau e^{i\Delta t + i\dot{\Delta}\tau - n C(t, \tau)}, \quad (9)$$

$$C(t, \tau) = \int d_r^3 d_v^3 f(v) \left[ 1 - e^{itV(r)/\hbar + i\tau \left( \frac{dV(r)}{dr} - v \right)/\hbar} \right].$$

Assuming for concreteness that all the dipoles are parallel to one another, we obtain from (9) a dispersion equation for  $W(\Delta)$  close to that obtained in [1]:

$$W(\Delta) = \frac{\Delta_0/\pi}{\Delta^2 + \Delta_0^2}, \quad \Delta_0 = \frac{2\pi\Omega_0}{9\sqrt{3}}. \quad (10)$$

Calculation of the joint distribution  $W(\Delta, \dot{\Delta})$  is very complicated. The distribution function for  $\dot{\Delta}$ , however, has a relatively simple near-dispersion form:

$$W(\dot{\Delta}) = \frac{1}{\pi} \int_0^\infty dx \cos\left(\frac{\dot{\Delta}x}{\dot{\Delta}_0}\right) e^{-x^{3/4}}, \quad \dot{\Delta}_0 = \frac{3}{4} (2\pi)^{1/3} n^{1/3} s \Omega_0. \quad (11)$$

The relaxation frequency can be defined as the reciprocal time of diffusion along the "energy axis"  $\hbar\Delta$ :

$$\gamma = \frac{\dot{\Delta}_0}{\Delta_0} = \frac{27\sqrt{3}}{4(2\pi)^{2/3}} n^{1/3} s. \quad (12)$$

Physically this result has the following meaning: The damping of the excitations is connected with the temporal fluctuations of the particles in an interaction sphere of radius  $r_0$ . In view of the fact that the interaction potential (1) decreases rapidly, these fluctu-

ations are determined by the nearest neighbor (pair collisions). Recognizing that the cross section of the pair collisions is in this case the square of the distance between particles ( $\sim n^{-2/3}$ ), and using the relation  $\gamma = n\sigma s$ , we obtain Eq. (12).

The author thanks V. L. Pokrovskii for a useful discussion.

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#### IRREVERSIBILITY OF TRANSITION OF NMR SIGNALS THROUGH A WEAK FIELD IN SOME MOLECULAR CRYSTALS

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ZhETF Pis'ma 5, No. 1, 16-17, 1 January 1967

Proton magnetic resonance signals in molecular crystals are characterized by a small spin-spin relaxation time ( $T_2 < 10^{-3}$  sec) and a large spin-lattice relaxation time ( $T_1 \geq 10$  sec). It is known [1,2,3] that the condition  $T_2 \ll T_1$  is necessary for the existence of a spin temperature in the nuclear-spin system of the sample. The existence of a spin temperature  $T_s(H)$  at arbitrary values of the constant field  $H$  was previously [1] demonstrated indirectly by means of an experiment wherein a sample was demagnetized in a weak (terrestrial) field: the sample in which the spin system has already assumed the lattice temperature was moved from the magnet gap to the terrestrial field and returned to the gap after a time  $t$  ( $T_2 \ll t \ll T_1$ ). No change in the signal was noted at all. The reversibility of the transition through a weak field served as one of the arguments in favor of the existence of a spin temperature  $T_s(H)$  in LiF crystals.

We performed a similar experiment with naphthalene single crystals. The transition through a weak field turned out to be irreversible for the NMR signal: a time  $t \approx 1$  turned out to be sufficient for total disorientation of the nuclear spins in these crystals; subsequent establishment of the equilibrium magnetization and a corresponding growth of the NMR signal occurred, as in the initial magnetization, with a time constant  $\tau \sim 10^3$  sec.

Neither variation of the intensity of the rf field  $H_1$  over a wide range, nor defects in the crystal lattice, have any influence on this effect. The behavior of the NMR signal in anthracene and biphenyl was similar.