Cooling of matter by a high frequency resonance field

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Several authors (Zel'dovich, 1974; Hansh and Schawlow, 1975; Shapiro, 1975), proceeding from various initial premises and employing different methods, have recently discussed the possibility of cooling a substance by means of high frequency resonant fields (under continuous action). This problem is further discussed in the present paper. The physical essence of cooling by means of periodic forces is analyzed. It is pointed out that there is a unity of the principles used in the various investigations. A number of examples of cooling a substance by an electromagnetic field are considered. It is shown, in particular, that strongly scattering but weakly absorbing media located in a multimode optical laser can be considerably cooled by a light beam (the efficiency is of the order of unity).

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1. INTRODUCTION

Scattering of high-frequency energy by a system usually heats the latter, but cooling is also possible under certain conditions. This is caused by the dichotomy of the scattering process. It is obvious that scattering is due to nonlinear interactions in the system. The energy-dissipation process is accompanied here by a redistribution of the thermal fluctuations over the frequency spectrum and over the motion modes inherent in the system. The resultant diequilibrium is interpreted in a number of cases as the appearance of subsystems with various temperatures. This disequilibrium leads to additional heat flow between the system and the thermostat, wherein motions with higher fluctuation levels, give up heat to the thermostat on the average, and those with lower fluctuation levels draw energy.

Imagine now that the coupling of the system with the external thermostat is not the same for the different frequencies or forms of motion. In practice, this is almost always the case. Then the rate of heat diversion to the thermostat via certain channels is not equal to the rate of heat flow from the thermostat via other channels. The resultant process is therefore a cooling of the system if the alternating forces produce in the system a disequilibrium that is appreciable in comparison with the direct dissipation and is of the required sign.

The thermodynamic picture is the following: the external forces, to maintain the thermal disequilibrium, perform work; the additionally scattered energy, together with the thermal energy of the system, goes to the thermostat. In the heater–evaporator–condenser–heater refrigeration cycle, the energy transport is connected with the displacement of the working medium. In the case analyzed here, on the other hand, energy is transferred continuously via temporal rather than spatial transformations.

One of the effects suitable for cooling is that of a source of monochromatic forces of frequency much higher than the characteristic frequencies of the thermal fluctuations in the system, but close to the frequencies of the resonances that interact nonlinearly with these fluctuations. This situation is favorable, for when the energy is pumped resonantly from the source to the rapidly oscillating subsystem, the energy exchange between the fast and slow motion becomes essentially unilateral: on one slope of the resonance curve the fluctuation energy flows only from the fast system to the slow one, and on the opposite slope the energy flow is reversed. This regular process is quite general in character and is dissipative.[4]

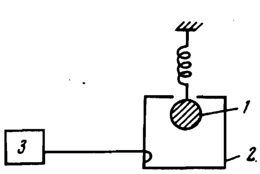
2. RESONATOR EXAMPLE AND ITS GENERALIZATION

By way of example, let us analyze the operation of a device (see the figure) that makes it possible to damp mechanical vibrations effectively by means of a high-frequency field. The body 1, which vibrates (say on a spring), forms together with cavity 2 an electromagnetic resonator of frequency ω_0. Electromagnetic power at a frequency ω close to ω_0 is fed from a source 3. The oscillations $x(t)$ of the body alter the frequency $\omega_0=\omega_0(x)$, and the ponderomotive forces of the field perform work on the body. If the generator 3 operates continuously, the mechanical vibrations of the body are damped at $\omega<\omega_0$. The excess energy is then absorbed by the resonator walls.

Let us estimate the rate of damping of the vibrations. The electric oscillations in the resonator are described by the equation

$$\ddot{q}+2\delta\dot{q}+\omega_0^2(x)q=a\cos\omega t, \tag{1}$$

where $\delta=\omega_0/2Q$, Q is the figure of merit of the resonator, and the quantity a is determined by the power delivered by the generator. If q is normalized so that $\frac{1}{2}(\dot{q}^2+\omega_0^2q^2)$ is the energy of the field in the resonator, then the sum of the ponderomotive forces acting on the body is



$$F_x = -\frac{1}{2} q^2 \frac{\partial}{\partial x} \omega_0^2.$$

At $\dot{x} \equiv 0$, forced oscillations of q, with frequency ω, take place. If a displacement $x(t)$ is produced, this regime changes, but with a delay relative to the changes of x. Consequently, work is performed during the $x(t)$ cycle. The power performed on the field in the case of small oscillations $x(t) \sim \varepsilon^{i\Omega t}$, with $\Omega \ll \omega$, averaged over the time $2\pi/\Omega$, amounts to

$$P(\Omega) = -\langle \dot{x} F_x \rangle = 2\langle \dot{x}^2 \rangle P_\omega \left(\frac{1}{\omega_0} \frac{\partial \omega_0}{\partial x} \right)^2 \frac{\omega_0^2}{\delta \Omega} S(\Omega), \quad (2)$$

$$S(\Omega) = \operatorname{Im} \left\{ \frac{1}{\Delta(\omega+\Omega)} - \frac{1}{\Delta(\omega-\Omega)} \right\}, \quad \Delta(\omega) = \omega_0^2 - \omega^2 + i2\delta\omega,$$

where $P_\omega = 2\delta \langle \dot{q}^2 \rangle = \delta \omega^2 a^2 / |\Delta(\omega)|^2$ is the power fed from the generator. If $\omega < \omega_0$, then at all Ω we have $S(\Omega)/\Omega > 0$, i.e., $P(\Omega) > 0$, meaning energy is continuously drawn from the source of the vibrations of x and is transformed into the high-frequency oscillations of q. The motions of x are damped. At $\omega > \omega_0$ the ratio is $S(\Omega)/\Omega < 0$, i.e., $P(\Omega) < 0$ and the reverse takes place—energy from the fast motions is transferred to the slow ones, and the motions of x tend to build up.

The flux P is appreciable in high-Q oscillating systems (small δ). At the same time, as $\delta \to 0$ the effect becomes weaker, for sooner or later a regime with $\delta \ll \Omega$ sets in and we then have an almost adiabatic, i.e., reversible change or passage through resonance. The effect vanishes also if the resonator is excited by a source that is not harmonic but has a spectrum that is broad in comparison with $|\omega - \omega_0| + \delta$.

The described method was developed[5] to suppress macroscopic vibrations. It is possible, however, to suppress also thermal vibrations of $x(t)$, i.e., Brownian vibrations. Let for example, body 1 together with the spring constitute a pendulum with frequency $\Omega_0 \ll \delta$. Then the spectrum of the $x(t)$ oscillations is concentrated in a frequency region $\ll \delta$ and we obtain for the rate of damping of the $x(t)$ fluctuations

$$P = \int_0^\infty P(\Omega)\, d\Omega \approx \langle \dot{x}^2 \rangle P_\omega \left(\frac{1}{\omega_0} \frac{\partial \omega_0}{\partial x} \right)^2 \frac{\omega_0}{\delta^3} \frac{y}{(1+y^2)^2},$$

where $y = (\omega_0^2 - \omega^2)/2\delta\omega$, $\langle \dot{x}^2 \rangle$ is the average intensity of the vibrations. At thermodynamic equilibrium we have $\langle \dot{x}^2 \rangle = kT/2M$, where M is the mass of the pendulum and kT is the Boltzmann factor. We have

$$\frac{1}{\omega_0} \frac{\partial \omega_0}{\partial x} \sim \frac{1}{L},$$

where L is the characteristic dimension of the field concentration of the field in the resonator. If we take a resonator with $L = 1$ cm, $\omega_0 = 10^8$ sec^{-1}, and $\delta = 10^4$ sec^{-1}, then at $M = 1$g, $P_\omega = 1$ W and at the best tuning ($y \approx 0.6$), an energy kT is transferred within $\sim 10^{-2}$ sec.

If the body 1 is a good conductor at the working frequencies of the resonator, or is a transparent dielectric, then it experiences no microwave heating. Furthermore, if the pendulum is in thermodynamic equilibrium at $P_\omega = 0$ with the field of the sources that cause the Brownian fluctuations $x(t)$, then the delivery of a power P_ω to the resonator initiates a lowering of the x fluctuation level, thus proving the feasibility of the cooling in principle. The flux of thermal energy from the body is initially equal to the flux P. With decreasing fluctuation intensity, the flux P decreases in proportion until it becomes weak enough to be commensurate with the power of the external random-force sources. It is this which determines the limiting cooling temperature. We note that one of the sources of the external forces is the thermal background of the electromagnetic radiation in the resonator.

It is easy to proceed from this example to a more general situation.

1. In the general case x is a multidimensional motion. It is convenient to represent it by a set of normal coordinates $x_1, x_2, \ldots$. Then

$$\frac{d\omega_0}{dt} = \frac{\partial \omega_0}{\partial x_1} \dot{x}_1 + \frac{\partial \omega_0}{\partial x_2} \dot{x}_2 + \ldots.$$

For independent coordinates, the small thermal fluctuations are independent, as follows from the Gibbs energy distribution; therefore

$$\left\langle \left(\frac{d\omega_0}{dt} \right)^2 \right\rangle = \left(\frac{\partial \omega_0}{\partial x_1} \right)^2 \langle \dot{x}_1^2 \rangle + \left(\frac{\partial \omega_0}{\partial x_2} \right)^2 \langle \dot{x}_2^2 \rangle + \ldots.$$

Each motion leaves its imprint on the response of the high-frequency resonance, and the combined heat flux is the sum of the fluxes from each low-frequency vibration mode.

2. A response is produced not only by the mechanical vibrations of the body, but also by its internal motions, if the latter lead to fluctuations of the electrical characteristic. The resultant random modulation of $\omega_0(t)$ causes an additional transfer of heat from the body 1 to the resonator. Let, for example, the body be a dielectric and let the dispersion region be far from the frequency ω. Then the electric losses are small and can turn out to be much lower than the pump power P transferred to the thermostat (to the resonator walls). Particularly pronounced are the fluctuations of the permittivity and permeability near the phase-transition points of the medium. The damping of critical fluctuations was discussed in[3], where the example used was a ferromagnet with a temperature close to the Curie point, placed in a microwave resonator.

3. As stressed in[3], the high frequency resonance can be produced not only by a resonant excitation system (resonator), but in the medium itself. If, for example, the thermal contact of the external thermostat with such resonances is much closer than the contact with the subsystem of low-frequency motions, then at $\omega < \omega_0$ the energy of the low-frequency fluctuations will be transformed upward in the spectrum and will leave the system.

We point in this connection to a communication by Hansh and Schawlow[2] (published simultaneously with[3]), in which it is proposed to quench thermal translational motions of gas molecules by exposing the gas to monochromatic light of frequency close to the absorption line of the gas. Hansh and Schawlow have noted the possibility of cooling the gas from 600 to 0.24 °K by this method. Their arguments and estimates were made in terms of the collisions of the gas molecules with the light quanta and were heuristic in character. In Sec. 3 we shall analyze this beautiful example again, but in terms of a vibrational approach, in order to cast light on a number of additional aspects of the phenomenon, which are of fundamental significance.

4. The capabilities of resonant cooling methods can be greatly increased by using a high-frequency oscillating system with a set of closely-spaced resonances. The transformation of the thermal energy takes place then as a result of random modulation of the resonance frequencies as well as of the parameters of the mutual coupling between the resonances and their coupling with the external source of frequency ω. We note that the power flux transferred to the thermostat can exceed the sum of the contributions of the changes $\langle (d\omega_k/dt)^2 \rangle$ for each of the high-frequency resonances ω_k. [3,4]

In Sec. 4 below we shall discuss a method for cooling a transparent (outside the absorption band) medium by placing it between the mirrors of a multimode optical resonator excited by a monochromatic beam. According to our estimates, this cooling method can be of practical interest.

5. We point out a connection between the foregoing and the well-known method of dynamic orientation of spins by a periodic field, and the associated change in the spin temperature, as reported by Zel'dovich. [1] In the analysis of this problem, one uses a model of a quantum-mechanical spin system constituting two interacting subsystems with partial-motion frequencies that differ by many orders of magnitude, the high-frequency two-level subsystem being resonantly pumped by an external monochromatic field. If the problem is analyzed in terms of the vibrational approach, then it turns out that the effect is based in essence on the same ability of the resonance to produce unidirectional energy flow from one subsystem to the other. It is important that if no damping is introduced in the system there will be no continuous energy transfer to the fast system. The quantization of the motions does not play a fundamental role here. We note also that cooling is produced also when the distribution of the fluctuations in the spin system can not be described by an effective temperature.

3. GAS IN A RESONANT FIELD

We shall adhere to the classical description; this is justified by the fact that we are interested in resonance in a sufficiently intense light field. We confine ourselves for simplicity to the case when the field intensity is much lower than the intensity that saturates the resonant transition. For the molecule-polarization oscillations we then have a model that is close to (1):

$$\ddot{q}+2\delta\dot{q}+\omega_0^2 q=a(x,\ t)=a_\omega^+ e^{i\omega(t+x/c)}+a_\omega^- e^{i\omega(t-x/c)}+\eta, \tag{3}$$

where $\eta(x,\ t)$ is a random thermal radiation field that is delta-correlated in t, $a_\omega^\pm$ are the amplitudes of the regular field of the oppositely directed waves, the frequency ω is close to the resonant frequency ω_0 of the transition, 2δ is the homogeneous width of the resonance ω_0, c is the speed of light, and $x=x(t)$ is the spatial position of the molecule (we confine ourselves for simplicity to the one-dimensional problem). In contrast to (1), the quantity fluctuating in (3) is not the frequency ω_0 but the frequency and amplitude of the external force. At $\dot{x}\equiv 0$ we have, against the thermal-background level, a regime of induced oscillations of q with frequency ω. In the case of Brownian motion of $x(t)$, this regime varies with a delay, and it is this delay which causes the unidirectional energy transfer from the motions of x to the radiation.

We choose for the sake of argument of coordinate q such that the quantity $(\dot{q}^2+\omega_0^2 q^2)/2$ represents the energy of the polarization oscillations (for an electric-dipole transition with unit oscillator strength we have in this case $a(x,\ t)=eEm^{-1/2}$, where e and m are the charge and mass of the electron, and $E(x,\ t)$ is the electric field strength). Then the force exerted on the molecule by the field is $F_x q\partial a/\partial x$, and the average flux density is

$$P_x=-\left\langle \dot{x}q\frac{\partial}{\partial x}a\right\rangle. \tag{4}$$

To calculate P_x we represent the functions $\cos(\nu x/c)$ and $\sin(\nu x/c)$ in the form

$$\cos\frac{\nu x}{c}=\int_{-\infty}^{\infty} e^{i\Omega t}\alpha_\nu(\Omega)\,d\Omega,\qquad \sin\frac{\nu x}{c}=\int_{-\infty}^{\infty} e^{i\Omega t}\beta_\nu(\Omega)\,d\Omega.$$

Then

$$q=\iint_{-\infty}^{\infty} e^{i(\nu+\Omega)t}H(\nu,\Omega)\frac{d\nu\,d\Omega}{\Delta(\nu+\Omega)},$$
$$H=(\alpha_\nu+i\beta_\nu)a_\nu^+ +(\alpha_\nu-i\beta_\nu)a_\nu^-,$$
$$a_\nu^\pm=a_\omega^\pm\delta(\nu-\omega)+(a_\omega^\pm)^*\delta(\nu+\omega)+\eta_\nu^\pm,$$

where $\delta(\omega)$ is the Dirac function, $\eta_\nu^\pm$ are the amplitudes of the expansion of $\eta(x,\ t)$ in terms of the traveling waves $\exp[i\nu(t\pm x)/c)]$. For the average power flux we get

$$P_x=-\iiiint_{-\infty}^{\infty}\exp[i(\nu+\nu_1+\Omega+\Omega_1)t]\langle H(\nu,\Omega)H(\nu_1,\Omega_1)\rangle\frac{i\Omega_1\,d\Omega_1\,d\Omega\,d\nu\,d\nu_1}{\Delta(\nu+\Omega)} \tag{5}$$

The correlation between the Brownian movement $x(t)$ and the fluctuating optical radiation is a weak secondary effect, and if it is neglected the white noise makes no contribution to P_x. For the case of a standing wave $a_\omega^+=a_\omega^-=a_\omega$, the contribution from the regular field is

$$P_x=|a_\omega|^2\int_{-\infty}^{\infty}\langle|\alpha_\omega|^2\rangle\frac{i\Omega\,d\Omega}{\Delta(\omega+\Omega)}+\text{c.c.} \tag{6a}$$

For a traveling wave (of the same intensity) we have

$$P_x=\frac{1}{2}|a_\omega|^2\int_{-\infty}^{\infty}\langle|\alpha_\omega|^2+|\beta_\omega|^2\rangle\frac{i\Omega\,d\Omega}{\Delta(\omega+\Omega)}+\text{c.c.} \tag{6b}$$

The gas is assumed to be rarefied enough to make the molecule mean free path $l \gg c/\omega$. We recall that $l = 1/\pi n\sigma^2$, where n is the gas density and σ is of the order of the molecule diameter ($l \sim 10^{-3}$ cm at $n = 10^{17}$ cm^{-3}). In the case of a gas of homogeneous density and $l \gg c/\omega$, using the result of Malakhov[6] for the shape of the $\cos\psi$ spectrum, when $\psi(t)$ is a Gaussian Markov process, we obtain

$$\langle|\alpha_\omega|^2\rangle = \langle|\beta_\omega|^2\rangle \approx \exp(-\Omega^2/u^2)/2\sqrt{\pi}\, u, \tag{7}$$

where $u^2 = 2\omega^2 kT/Mc^2$ and M is the molecule mass.

Substitution in (6) yields

$$P_x = |a_\omega|^2 A(u)/4\omega_0, \tag{8}$$

$$A(u) = z_+ Z(z_+) - z_- Z(z_-) + \text{c.c.}, \qquad Z(z) = \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} e^{-s^2} \frac{ds}{s - iz},$$

where $z_\pm = [\delta + i(\omega \pm \omega_0)]/u$ and $Z(z)$ is a tabulated function known from gas optics.[7]

If the gas temperature is not too low, then $u \gg \delta$ (but $u^2/\omega^2 = 2kT/Mc^2 \ll 1$). Then

$$A(u) \approx 2\sqrt{\pi}\frac{\omega_0 - \omega}{u} \quad \text{if} \quad |\omega_0 - \omega| \ll u, \tag{9a}$$

$$A(u) \approx \frac{4u^2\delta(\omega_0 - \omega)}{[(\omega_0 - \omega)^2 + \delta^2]^2} \quad \text{if} \quad |\omega_0 - \omega| \gg u. \tag{9b}$$

In the region of optimal tuning ($\omega_0 - \omega \sim u$) we have $A \sim 1$, i.e., P_x is independent of the density and temperâture of the gas. At $\omega = 10^{16}$ sec^{-1}, $\delta = 10^9$ sec^{-1}, and $M = 10^{-22}$ g, the values of u and δ become comparable at $T \sim 3$ °K. In the case of deep cooling, when $u \ll \delta$, the quantity A behaves in accordance with relation (9b) at all detunings if its maximum value is $A \sim u^2/\delta^2 \ll 1$.

We present an estimate of $|a_\omega|^2/4\omega_0$. For a transition with an oscillator strength f we have

$$\frac{|a_\omega|^2}{4\omega_0} = f^2 \frac{\pi e^2}{mc}\frac{p}{\omega_0},$$

where $p = c\overline{E^2}/8\pi$ is the light flux of frequency ω, per unit area, with $\pi e^2/mc = 0.0265$ cm^2/sec. At $\omega = 10^{16}$ sec^{-1} and $p = 10^{-3}$ W/cm^2 ($E \sim 1$ V/cm) we have $\pi e^2 p/mc\omega_0 \sim 2 \cdot 10^{-14}$ erg/sec, corresponding, at $f = 1$ and at the best tuning, to cooling of the x-motions of the molecules at a rate $\sim 10^2$ deg/sec. On the other hand if we substitute $p = 10^3$ W/cm^2 (this is the flux used by Hansh and Schawlow), we obtain a cooling rate of 1000 degrees in $\sim 10^{-5}$ sec.

It is interesting that this estimate coincides with that given by Hansh and Schawlow,[2] although it was obtained by them from other considerations, using data on the radiative line width, the molecule mass, the gas temperature, and the field strength required to saturate the resonant transitions. Our final result, on the other hand, contains besides the condition of optimal tuning only one parameter—the oscillator strength. We note that a flux of 10^3 W/cm^2 may turn out to be large, in the sense of saturation of the transition, when the model (3) linear in q ceases to be valid.

The total flux of the x fluctuation power converted into radiation, in the case of a rarefied gas, is determined by the quantity P_x multiplied by the molecule concentration.

Besides damping the translational motions of the gas molecules (the only motion considered in[2]), there is also damping of other modes of low-frequency vibrations of molecules, if they influence the resonance parameters q (we recall Sec. 2.2). These vibrations are, for example, the rotational motions of the molecules, since the optical polarizability of the molecules is practically never isotropic. The effect is easy to account for within the framework of the model (3), by assuming that a depends not only on x but also on the molecule orientation angles. In the case of Brownian rotational motions of the molecule, additional random modulation of the right-hand side of (3) takes place, and by virtue of the delayed reaction of the regime of the q-oscillations to the rotations, the rotation energy will be converted into radiation in exactly the same manner as in the case of fluctuations of x.

Let, for example, the molecule have a strongly pronounced polarizability along one direction. Then allowance for the rotation reduces to multiplication of the field amplitudes by $\cos\varphi$, where $\varphi(t)$ is the angle between the direction of the electric wave vector and the selected molecule axis. We now have in lieu of (4)

$$P = P_x + P_\varphi = -\left\langle \dot{x} q \frac{\partial}{\partial x} a \right\rangle - \left\langle \dot{\varphi} q \frac{\partial}{\partial \varphi} a \right\rangle.$$

If the field of the regular wave is linearly polarized, then φ is the molecule orientation angle in the laboratory frame. Representing $\cos\varphi$ in the spectral form

$$\cos\varphi = \int_{-\infty}^{\infty} e^{i\Omega t}\gamma(\Omega)\, d\Omega$$

and taking into account the statistical independence of the $x(t)$ and $\varphi(t)$ fluctuations, we obtain in analogy with (6)

$$P_x = |a_\omega|^2 \iint_{-\infty}^{\infty} \langle|\alpha_\omega(\Omega)|^2\rangle\langle|\gamma(\Omega_1)|^2\rangle \frac{i\Omega\, d\Omega\, d\Omega_1}{\Delta(\omega + \Omega + \Omega_1)} + \text{c.c.},$$

$$P_\varphi = |a_\omega|^2 \iint_{-\infty}^{\infty} \langle|\alpha_\omega(\Omega)|^2\rangle\langle|\gamma(\Omega_1)|^2\rangle \frac{i\Omega_1\, d\Omega\, d\Omega_1}{\Delta(\omega + \Omega + \Omega_1)} + \text{c.c.},$$

We assume in the estimates that $\langle\dot{\varphi}^2\rangle = 2kT/J$, where J is the moment of inertia of the molecule; this is correct at $T \gg T_\varphi = h^2/8\pi^2 Jk$, where h is Planck's constant (T_φ is usually of the order of several units or several fractions of a degree Kelvin). The $\varphi(t)$ fluctuation spectrum falls off at frequencies lower than or of the order of $\Omega_* = l^{-1}(kT/M)^{1/2}$. Recognizing that $J < M\sigma^2$, we have

$$\frac{\langle\dot{\varphi}^2\rangle}{\Omega_*^2} \geqslant \frac{2kT}{J}\frac{Ml^2}{kT} > \frac{l^2}{\sigma^2} \gg 1.$$

We therefore obtain for the fluctuation spectrum of $\cos\varphi$, in analogy with (7)

$$\langle|\gamma(\Omega)|^2\rangle \approx \frac{1}{2\sqrt{\pi}\, w}\exp\left(-\frac{\Omega^2}{w^2}\right), \qquad w^2 = 2\langle\dot{\varphi}^2\rangle = \frac{4kT}{J}. \tag{10}$$

Since

$$\frac{w^2}{u^2}=\frac{4kT}{J}\frac{Mc^2}{2\omega^2kT}>\frac{2c^2}{\omega^2\sigma^2}\gg 1,$$

it follows that the rate of damping of the rotational motions is approximately

$$P_\varphi\approx|a_\omega|^2A(w)/4\omega_0,$$

and the damping of the x motion is sharply weakened in comparison with (8). Now P (P_x and P_φ) is maximal at $\omega_0-\omega\sim w$, i.e., at detunings greatly exceeding the Doppler width u. At the maximum we have $P_\varphi\sim|a_\omega|^2/4\omega_0$, and the ratio $(P_x/P_\varphi)_{\max}\lesssim u/\omega\sim 10^{-2}$.

Similar manipulations can be performed also for the case of an elliptically polarized wave a_ω.

The background of the radiation scattered by the molecules was not taken into account in the foregoing (the field of a_ω was assumed given). It is obvious that it cannot play any role as $n\to 0$, since the cooling rate per molecule P, does not depend on n or T at $u\gg\delta$ or at $w\gg\delta$ and at optimal tuning, and is equal to $|a_\omega|^2/4\omega_0$. Nor did we take into account the heat transferred to the x and φ motions from the "hot" modes of motion. These are, in particular, the oscillations of the polarization (of frequency φ_0). The collisions bring about a rather rapid energy exchange between the molecule vibrations whose frequencies are commensurate with ω_0. These modes of motion become heated. However, once the molecules leave the irradiation zone (or once the field a_ω is turned off), the energy of these motions is radiated much more rapidly than the heat transfer to the x and φ motions, since nonradiative transitions in a rarefied gas are negligibly weak at normal and low temperatures. This ensures (disregarding the walls) the possibility of cooling the gas.

To what temperatures can the gas be cooled? Hansh and Schawlow[2] connect this limit with the condition $u\sim\delta_0$, where δ_0 is the natural line width ($\delta_0=\lim_{n\to 0}\delta$). According to calculation $P\neq 0$ for all T, but as $T\to 0$ a regime with $u\ll\delta$ and $w\ll\delta$ sets in, and the rate of cooling falls off like u^2/δ^2 or w^2/δ^2. The cooling limit is determined, on the one hand, by the decrease of P, and on the other by the increase of the heat transfer from the hot motions. In principle, therefore, the limit in T can correspond also to $u\ll\delta_0$. We note that as the gas becomes cooled relative to the background temperature η, this background also begins to contribute to the heating. In our model (3) this corresponds to the appearance at $a_\omega\neq 0$, of a correlation $\langle H(\nu,\Omega)H(\nu_1,\Omega_1)\rangle$, which was neglected when the results (6) were obtained from (5). Since the particle collisions violate this correlation, it manifests itself stronger the smaller $u/(\delta-\delta_0)$. The smallness of $u/(\delta-\delta_0)$, however, does not exclude the possibility of $u\ll\delta_0$.

When estimating the limiting temperature one must also take into account the possibility of quenching of various motions that are quantized. In particular, the rotational motions are quantized. There is a factor (which we have purposefully disregarded in the preceding analysis), that gives rise to an additional quenching of the x and φ motions. The point is that in the derivation of (7) and (10) the gas was assumed to be homogeneous and isotropic. But since $a=a(x,\varphi)$, the time-averaged force $\langle F_x\rangle$ acting on the molecule and the torque $\langle F_\varphi\rangle=\langle q\partial a/\partial\varphi\rangle$ are both functions of x and φ. For low-frequency ($\ll\delta$) motions of x and φ in the field of a linearly polarized standing wave we have

$$\langle F_x\rangle\approx-8|a_\omega|^2\frac{\omega}{c}\frac{\omega_0^2-\omega^2}{|\Delta(\omega)|^2}\sin\frac{2\omega x}{c}\cos^2\varphi,$$

$$\langle F_\varphi\rangle\approx-8|a_\omega|^2\frac{\omega_0^2-\omega^2}{|\Delta(\omega)|^2}\cos^2\frac{\omega x}{c}\sin 2\varphi.$$

At $\omega_0>\omega$ the molecules tend to align themselves along the field ($\varphi=0$) and to land in its antinode ($x=0$).[1)] An important role is played in this problem by the homogeneity of the gas at distances $x\sim c/\omega$ and its isotropy at angles $\varphi\sim 1$. The potential energy corresponding to these deviations is

$$\Pi\approx4|a_\omega|^2\frac{\omega_0^2-\omega^2}{|\Delta(\omega)|^2}\leqslant\frac{|a_\omega|^2}{2\delta\omega}.$$

If the incident light flux has a high intensity, so that Π exceeds kT, then the gas becomes inhomogeneous and anisotropic, the intensities of the x and φ fluctuations decrease, and the cooling effect becomes weaker. At $\omega=10^{16}\ \sec^{-1}$, $\delta=10^9\ \sec^{-1}$, $p=10^3\ \mathrm{W/cm^2}$, and $f=1$ the value of $|a_\omega|^2/2\delta\omega$ becomes comparable with kT at $T\sim 0.4\,°\mathrm{K}$.

We note that Hansh and Schawlow have proposed to cool the gas by isotropic irradiation, i.e., by opposing light fluxes. However, all the estimates (including the estimate of the potential Π) are practically the same for a standing wave as for a traveling wave.

4. TRANSPARENT MATTER IN MULTIMODE OPTICAL RESONATOR

We consider a situation wherein the light flux has a low intensity, so that w in estimates of the work performed over short times the connection between the polarization of the medium and the field can be regarded as linear. Let the connection be local. Far from the absorption bands it can then be described by a real permittivity tensor

$$\varepsilon=\varepsilon_0+\varepsilon_\sim(\mathbf{r},t),\quad\varepsilon_0=\langle\varepsilon\rangle.$$

We expand the electric field $\mathbf{E}(\mathbf{r},t)$ in a resonator filled with a medium in terms of the set $\{\mathbf{E}_k(\mathbf{r})\}$ representing the normal modes in the system at $\varepsilon\equiv\varepsilon_0$:

$$\mathbf{E}(\mathbf{r},t)=\sum_k q_k(t)\mathbf{E}_k(\mathbf{r}).$$

We use the normalization

$$\frac{1}{4\pi}\int\mathbf{E}_k\varepsilon_0\mathbf{E}_n\,d\mathbf{r}=\delta_{kn}$$

where δ_{kn} is the Kronecker symbol and the integration is over the volume V of the resonator. Maxwell's equations take in this representation the form

$$\ddot{q}_k+\delta_k\dot{q}_k+\omega_k^2 q_k=\sum \frac{d^2}{dt^2}(a_{kn}q_n)+f_k+f_k^{(c)}, \qquad (11)$$

$$a_{kn}=-\frac{1}{4\pi}\int \mathbf{E}_k\varepsilon_\sim \mathbf{E}_n\,d\mathbf{r},$$

where ω_k is the natural frequency of the mode k, the parameters δ_k take into account the absorption in the mirrors and the diffraction losses, $f_k(t)$ are the sources of the regular excitation of the mode k and are determined by the intensity and geometry of the beam incident on the mirror from the outside, and $f_k^{(c)}$ are the sources of the thermal excitation and their intensity is such that at $f_k\equiv 0$ they cancel the energy outflow $\sim\delta_k\langle\dot{q}_k^2\rangle$, so that thermodynamic equilibrium obtains in the system. The random process $f_k^{(c)}$ can be regarded as δ-correlated in the time t.

When the resonator is illuminated with light of frequency ω we have $f_k=f_{0k}e^{i\omega t}+\text{c.c.}$ We consider the forced oscillations of $q(t)$ produced above the level of the thermal background. We denote by $\{q_{01}, q_{02}, \ldots\}$ the amplitudes of the forced oscillations of frequency ω at $\varepsilon\equiv\varepsilon_0$. The time fluctuations of ε lead to modulation of this regime. Owing to the finite Q of the resonances (δ_k is finite), the electric regime varies with a delay relative to the variation of $\varepsilon(t)$. It is this which makes it possible for the work to differ from zero in one cycle of a periodic (random) variation of $\varepsilon_\sim$. The work per unit time is on the average

$$P=-\frac{1}{8\pi}\int\langle\mathbf{E}\dot{\varepsilon}\mathbf{E}\rangle d\mathbf{r}=\frac{1}{2}\sum_{k,n}\langle q_k q_n \dot{a}_{kn}\rangle. \qquad (12)$$

At $P>0$, the fluctuation energy is transferred from the medium to the radiation, and is then dissipated because δ_k is finite, so that the medium is cooled. At $P<0$ the effect is reversed and the medium is heated.

We estimate P by starting from (11) and (12). We represent $\varepsilon_\sim$ in the spectral form

$$\varepsilon_\sim(\mathbf{r},t)=\int_{-\infty}^{\infty}\varepsilon(\mathbf{r},\Omega)e^{i\Omega t}d\Omega,$$

and obtain from (11) and (12), accurate to terms quadratic in the small $\varepsilon_\sim$ and f,

$$P=\sum_k\int_{-\infty}\left[\frac{i\Omega(\omega+\Omega)^2}{\omega_k^2-(\omega+\Omega)^2+i(\omega+\Omega)\delta_k}+\text{c.c.}\right]B_k\,d\Omega, \qquad (13)$$

$$B_k(\Omega)=\sum_{m,n} q_{0m}q_{0n}^*\langle a_{mk}(\Omega)a_{kn}^*(\Omega)\rangle.$$

In this calculation, the fluctuations of $\varepsilon_\sim$ and $f^{(c)}$ were assumed to be uncorrelated. Such a model is suitable for the investigation of the rate of the initial cooling stage, to which we confine ourselves.[2)]

Inasmuch as $\hbar\omega\gg kT$ for optical frequencies ω and ordinary temperatures T, it follows that the intensity of the thermodynamic fluctuations of $\varepsilon_\sim$, meaning also the $B_k(\Omega)$ spectrum, decreases exponentially at frequencies much lower than ω. Recognizing also that $B_k(\Omega)=B_k(-\Omega)>0$, it is easy to verify that resonances $\omega_k>\omega$ make positive contributions to P, and $\omega_k<\omega$ negative ones. A small contribution is made by high-Q resonator modes for which $|\omega_k-\omega|\lesssim\Omega_s$ and $\delta_k\lesssim\Omega_s$, where Ω_s is the characteristic frequency of the decrease of B_k. Consequently, to attain the cooling effect the spectrum of the high-Q resonances $\{\omega_k\}$ must be dense in the band $(\omega, \omega+\Omega_s)$ and be widely spaced (or nonexistent) in the band $(\omega-\Omega_s, \omega)$.

We assume next that: 1) the resonator dimensions are large both in comparison with λ and in comparison with the range of the excitations of the medium that make the main contribution to the light scattering,[3)] and 2) that $f_{0k}=f_0\delta_{0k}$, i.e., the external beam excites only one resonator mode with index 0. The first condition makes it possible to approximate the wave field $\mathbf{E}_k(\mathbf{r})$ by a three-dimensional harmonic structure, this being equivalent to superposition of four standing waves with wave vectors $\mathbf{k}_i(k)$, $i=1, 2, 3, 4$. Thus, a_{kn} is a sum of 64 terms, each proportional to $\varepsilon(\varkappa, \Omega)$, with

$$\varepsilon(\varkappa,\Omega)=\frac{1}{V}\int\varepsilon(\mathbf{r},\Omega)e^{i\varkappa\mathbf{r}}d\mathbf{r}$$

with $\varkappa$ from the set $\varkappa(k, n)=\pm\mathbf{k}_i(k)\pm\mathbf{k}_j(n)$; $i, j=1, 2, 3, 4$. By virtue of condition 2) we have

$$B_k=|q_0|^2\langle|a_{0k}|^2\rangle=\frac{1}{2\delta_0}P\sum_{\varkappa(0,k)}d_\varkappa\langle|\varepsilon(\varkappa,\Omega)|^2\rangle, \qquad (14)$$

$$P_0=\frac{2\delta_0|f_0|^2}{(\omega_0^2-\omega^2)^2+\omega^2\delta_0^2},$$

where P_0 is the power fed to the resonator from the source, while the parameters $d_\varkappa$ are determined by the symmetries of ε_0 and $\varepsilon_\sim$ and of $\mathbf{E}_0(\mathbf{r})$ and $\mathbf{E}_k(\mathbf{r})$. It is taken into account in (14) that $\langle\varepsilon(\varkappa,\Omega)\varepsilon(\varkappa_1,\Omega)\rangle\neq 0$ only at $\varkappa+\varkappa_1=0$. Inasmuch as only oscillations with $|\omega_k-\omega|\lesssim\Omega_s\ll\omega$ are significant in (13), it can be assumed that $|\varkappa|\lesssim 4\pi/\lambda$.

When considering the contribution to $\varepsilon_\sim$ from various modes of motions of the medium, two limiting cases can be distinguished. In one case the excitations are propagated in wavelike manner (e.g., elastic Debye waves), with

$$\langle|\varepsilon(\varkappa,\Omega)|^2\rangle=\frac{2}{\pi}\langle|\varepsilon_1(\varkappa)|^2\rangle\frac{\Omega_\varkappa^2\delta_\varkappa}{(\Omega_\varkappa^2-\Omega^2)^2+4\Omega^2\delta_\varkappa^2}, \qquad (15a)$$

$\Omega_\varkappa=\Omega(\varkappa)$ is the dispersion law, and $\delta_\varkappa=\delta(\varkappa)$ is the wave damping decrement. In the second case the excitations propagate in diffuse manner, with

$$\langle|\varepsilon(\varkappa,\Omega)|^2\rangle=\frac{1}{\pi}\langle|\varepsilon_2(\varkappa)|^2\rangle\frac{\sigma_\varkappa}{\Omega^2+\sigma_\varkappa^2}, \qquad (15b)$$

$\sigma_\varkappa=\chi\varkappa^2$, and χ is the diffusion coefficient. A contribution in the form (15b) is made to $\varepsilon_\sim$, for example, by fluctuations of the entropy or of the chemical potential. This part of $\varepsilon_\sim$ determines the Rayleigh scattering of the light. The part in the form (15a) is due to Mandel'shtam-Brillouin scattering and to Raman scattering (for more details on the $\varepsilon_\sim$ spectra see, e.g., Fabelinskii's book[8]).

We substitute (14) and (15) in (13) and integrate with respect to Ω, recognizing that the only essential oscillations are those for which δ_k, $\Omega_\varkappa$, $\delta_\varkappa$, and $\sigma_\varkappa$ are much smaller than ω. For the spectrum (15a) we obtain at $\Omega \gg \delta_\varkappa$

$$\frac{P_1}{P_0} \approx \sum_k \sum_{\varkappa(0,k)} \langle |\varepsilon_1(\varkappa)|^2 \rangle \frac{d_\varkappa g_1}{2\delta_0} \left[\frac{\omega_+^2 \Omega_\varkappa}{(\omega_k^2-\omega_+^2)^2+\omega_+^2 g_1^2} - \frac{\omega_-^2 \Omega_\varkappa}{(\omega_k^2-\omega_-^2)^2+\omega_-^2 g_1^2} \right],$$

and for the spectrum (15b)

$$\frac{P_2}{P_0} \approx \sum_k \sum_{\varkappa(0,k)} \langle |\varepsilon_2(\varkappa)|^2 \rangle \frac{d_\varkappa \sigma_\varkappa}{\delta_0} \frac{\omega^2(\omega_k^2-\omega^2)}{(\omega_k^2-\omega^2)^2+\omega^2 g_2^2},$$

where $\omega_\pm = \omega \pm \Omega_\varkappa$, $g_1 = \delta_k + 2\delta_\varkappa$, and $g_2 = \delta_k + 2\sigma_\varkappa$. We note that $\varkappa(0, k)$ and $d_\varkappa$ are quite irregular functions of $\mathbf{k}(k)$. Therefore if a set of resonances $\{\omega_k\}$ with close values of δ_k exists and there is no sharp anisotropy in the system (i.e., in ε_0 and in the distribution of the wave vectors $\mathbf{k}(k)$), then we can regard $\varkappa$ in the double sums approximately as a parameter independent of k, which is continuously distributed in the interval $|\varkappa| \leq 4\pi/\lambda$, and we can assume $d_\varkappa = 1/64\,\varepsilon_0^2$.

Let us estimate the contribution made to P by the isotropic part of $\varepsilon_\sim$ for the case of an isotropic medium. We assume that $\langle|\varepsilon(\varkappa)|^2\rangle \equiv \langle\varepsilon_\sim^2\rangle$ (over scales $|\varkappa| \leq 4\pi/\lambda$, the dependence of ε on $\varkappa$ is usually significant only at critical points of the thermodynamic state of the medium). Then, as can be easily verified,

$$\langle \varepsilon_\sim^2 \rangle = \frac{3}{8\pi^3} \frac{\lambda^4 H}{V},$$

where H is the extinction coefficient (more accurately, its part due to isotropic fluctuations of $\varepsilon_\sim$).

In the most favorable case, when all the resonances of the resonator of frequencies $\omega_k < \omega$ have in a band $\delta_\varkappa \gg \Omega_s$ close to ω a low Q, $\delta_k \gg \Omega_s = kT/\hbar$, and the density of the high-Q resonances $\omega_k > \omega$ with $\delta_k \lesssim \Omega_s$ is high, we obtain

$$\frac{P_1}{P_0} \approx \mu H_1 \frac{\lambda}{\varepsilon_0^2} \frac{\langle\Omega^2\rangle}{\delta_0 \omega}, \quad \frac{P_2}{P_0} \approx \mu H_2 \frac{\lambda}{\varepsilon_0^2} \frac{\sigma}{\delta_0} \ln\frac{\omega}{g_2}, \tag{16}$$

where $H_{1,2}$ is the part of H due to the corresponding type of motion of the medium, $\mu = N/N_0$, $N_0 = 8\pi V/\omega\lambda^3$ is the number of all the oscillations of the volume V in a unit frequency interval, and N is the density of the high-Q modes for which $\delta_k \lesssim \Omega_s$. The quantity $\langle\Omega^2\rangle$ is the average of $\Omega_\varkappa^2$ over $\varkappa$ in the interval $|\varkappa| < 4\pi/\lambda$ and $\langle\sigma \ln(\omega/g_2)\rangle$ is averaged both over $\varkappa$ and over the distribution of δ_k. In the estimates it is assumed that the resonances $\{\omega_k\}$ with close values of δ fill uniformly a broad ($> \delta$) frequency band.

We present numerical estimates, assuming $\omega_0/\delta_0 \sim \omega_k/\delta_k \sim 10^{10}$ and $\mu \sim 1$. We take by way of example the medium to be pure air under normal conditions. The molecule mean free path in it is $l \sim 10^{-5}$ cm, i.e., $\lambda/l \sim 5$, and the medium can be approximately regarded as continuous and the dispersion neglected. According to the handbook data (see e.g., [8]), the main contribution to H is made by scattering from the adiabatic ($\sim H_1$) and isobaric ($\sim H_2$) density fluctuations. We take into account the fact that $\Omega^2/\omega^2 \sim v^2/c^2 \sim 10^{-10}$, where v is the speed of sound, and that

$$\left\langle \frac{\sigma}{\delta_0} \ln\frac{\omega}{g_2} \right\rangle \sim \frac{\langle\sigma\rangle}{\delta_0} \ln\frac{\omega}{2\langle\sigma\rangle} \sim 10^{-6}$$

(since $\langle\sigma\rangle \sim \chi\omega^2/c^2$ and $\chi \approx 0.2$ cm^2/sec). For air the total value is $H \sim 3\times10^{-7}$ cm^{-1} (at $\lambda = 4350$ Å). It follows from the Landau-Placzek formula that

$$H_2/H_1 = (c_p - c_v)/c_v,$$

where c_p and c_v are the specific heats at constant pressure and volume, and for air $(c_p - c_v)/c_v \approx 0.4$. We thus obtain

$$P_1/P_0 \sim 10^{-11}, \quad P_2/P_0 \sim 10^{-5}.$$

If the medium chosen is a pure liquid such as benzene, then we have $H \approx 10^{-3}$ cm^{-1} and, assuming that the contribution to H due to density fluctuations is of the order of H, we obtain the estimates

$$P_1/P_0 \sim 10^{-6}, \quad P_2/P_0 \sim 10^{-4}.$$

For solutions of polymers and proteins, $H \sim 10^{-1}$ cm^{-1} and can be even larger; P_1 and P_2 increase correspondingly.

We note that part of the scattering in the media is connected with thermal excitations not of the acoustic modes but of the activation modes of the vibrations of the medium, for which $\langle\Omega^2\rangle \gg \omega^2 v^2/c^2$. For examples, for the above-mentioned liquids an appreciable contribution to H_1 is made by the conformation vibrations of the molecules with frequencies $\Omega \sim 10^{11}-10^{13}$ sec^{-1}. Another example is provided by transparent ferroelectrics with large electro-optical constants, in which the greater fraction of the scattering is due to polarization oscillations with frequencies of the same order. We note that we must have $\Omega < \Omega_s$. At $T = 100$ °K we have $\Omega_s^2/\omega^2 \sim 2\times10^{-3}$. At $H_1 \sim 10^{-2}$ cm^{-1} and $\langle\Omega^2\rangle/\omega^2 \sim 10^{-5}$ we obtain $P_1/P_0 \sim 1$.

The foregoing estimates show that the cooling effect can be quite appreciable for weakly absorbing but strongly scattering media. We recall that the calculation was carried out for an ideally nonabsorbing medium. At a finite but small absorption it is necessary to replace δ_k in the calculations by $\delta_k + \omega\lambda\!\!\!^{-} D$, where $\lambda\!\!\!^{-} = \lambda/2\pi$ and D is the absorption coefficient (the light intensity is extinguished by a factor 2.7 over a length D^{-1}). The Joule heating power of the medium amounts to

$$P_D = P_0 \frac{\omega\lambda\!\!\!^{-} D}{(\delta_0^2 + \omega^2\lambda\!\!\!^{-2} D^2)^{1/2}}.$$

The cooling will obviously take place if P exceeds P_D. Under conditions when H_1 or H_2 prevails, this means that for the cooling we should have

$$\frac{D}{H_1} < \frac{2\pi\mu\langle\Omega^2\rangle}{\varepsilon_0^2\omega^2} \text{ или } \frac{D}{H_2} < \frac{2\pi\mu}{\varepsilon_0^2} \left\langle \frac{\sigma}{\omega} \ln\frac{\omega}{g_2} \right\rangle.$$

We have no data on the values of D for media with large H. It follows from the physical considerations mentioned in Sec. 3 that the requirement that the absorption be small in comparison with the scattering can in all cases be satisfied for gases.

5. CONCLUSION

Thus, the cooling of matter by a high-frequency field, which seems paradoxical at first glance, does indeed follow from rather elementary considerations. The effect lends itself to experimental observation and one might think also to applications.

In this paper the cooling problem was considered under conditions of a relatively weak external alternating field. With increasing field, various unaccounted-for nonlinear processes in the system will come into play (in particular, the nonlinearity of Eqs. (1), (3), and (11) with respect to q). It is important that higher-order nonlinearities can either weaken or enhance the cooling effect (depending on the sign of the anharmonicity). We note also (see [4] on this subject) that unidirectional energy fluxes are produced not only at the fundamental resonance $\omega \sim \omega_k$, but also in nonlinear resonances of higher orders (e.g., at the parametric resonance $\omega \sim 2\omega_k$).

1)The quenching of slow motions in high-frequency resonances is always accompanied by changes in the effective rigidity of these motions. For the examples discussed in the text, the changes in the susceptibilities of the slow motions are connected by the Kramers-Kronig relations.

2)At $f_k \equiv 0$ in the thermodynamic state, the average work above the thermal radiation background at fluctuations of ϵ_- is always equal to zero. It can be disregarded also in the case of small deviations from this regime. On the other hand if $f_k(t)$ causes accumulation of a finite difference between the temperatures of the medium and of the radiation (i.e., the temperature of the resonator walls), then work is performed on the average, i.e., a correlation appears between ϵ_- and $f^{(c)}$. The resultant heat flux ultimately balances the flux (13); it is this which determines the establishment of the stationary temperature regime of the system.

3)Excitations with large free paths usually fluctuate slowly in time, i.e., they contribute to $B_k(\Omega)$ at $\Omega \approx 0$. However, as $\Omega \to 0 (\Omega \ll \delta_k)$ the factor preceding B_k in (13) is small, so that the contribution to P from these fluctuations is negligible.

4)We recall that H^{-1} is the thickness of the medium over which the light intensity is attenuated by the scattering by a factor 2.7.

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Photoelectric and acousto-electric fields in superconductors

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If a superconductor is exposed to microwave radiation or if sound propagates through a superconductor, stationary electric fields arise in such a superconductor and decrease with the distance from the boundary. We obtain equations which describe the distribution of these fields and the boundary conditions for them. We discuss methods of observation and find the correction to the frequency of the Josephson radiation if one of the superconductors is irradiated by uhf radiation or sound.

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1. INTRODUCTION

We study in the present paper how electric fields arise in a superconductor under the action of microwave radiation or longitudinal sound. Rieger, Scalapino, and Mercereau[1] and Tinkham and Clarke[2] were the first to indicate the possibility that there might exist stationary electric fields in superconductors. They showed that if a current passes through a S–I–N contact, electric fields arise in the superconductor which decrease far from the boundary at a diffusion distance $L^2 = D\tau_Q$ in superconductors with a gap[2] and at a coherence length in gapless superconductors.[1] The time τ_Q of the relaxation of the excitations between two branches of the spectrum $\xi_p > 0$ and $\xi_p < 0$ ($\xi_p = p^2/2m - \mu$, $\mathbf{p}$ is the quasi-momentum of the electrons, μ the chemical potential of the normal metal, and D the diffusion coefficient of the normal electrons) caused, for in-

