

# Laser cooling of impurity crystals

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**Abstract.** A review of the main experimental achievements in the new field of physics – laser cooling of solids – is presented. The method of nonequilibrium statistical operator is used to develop the theory of laser cooling of an impurity crystal. Predictions of this theory are compared with the results of experiments carried out at Los Alamos National Laboratory (USA).

**Keywords:** impurity crystals, laser cooling, nonequilibrium statistical operator

## 1. Introduction

An impressive progress has been recently made in the laser cooling of gas media. In particular, Bose–Einstein condensation of matter was achieved through laser cooling. In 1997, Chu, Cohen-Tannoudji, and Phillips were awarded the Nobel Prize in physics for their research into the laser cooling of gas media.

The possibility of optical cooling of continua was predicted by Pringsheim long time ago [1]. This prediction initiated a theoretical discussion between Pringsheim [2] and Vavilov [3, 4], whose thermodynamic arguments cast doubt on the possibility of laser cooling of continuum. This controversy came to an end when Landau [5] developed a consistent thermodynamic theory proving the possibility of optical cooling of continua.

However, because of a complex structure of continua, the optical cooling of such media was observed only in recent experiments [6, 7], using tunable narrow-band lasers and ultrahigh-purity media.

The theory of laser cooling of impurity molecular crystals [8, 9] and pure molecular crystals [10] has been developed by Andrianov and Samartsev. Oraevskii [11] and Zadernovskii and Rivlin [12] have developed the theory of laser cooling of semiconductors.

In this paper, we present a more detailed theory of laser cooling of impurity crystals, including the spatial inhomogeneity of phonon states.

The inclusion of this factor may be important for the creation of a practical laser refrigerator, when the inhomogeneity of the phonon flux near the boundaries of a sample and the nonuniformity of its internal structure have to be taken into account. We will also examine the case of phonons with an arbitrary dispersion.

## 2. Experimental

The first experiment on the real cooling of a solid sample was performed by Epstein et al. at Los Alamos National Laboratory (USA) in 1995 [6]. In this experiment a room-temperature sample of heavy metal–fluoride glass doped with ytterbium ions was cooled by 0.3 K through the saturation of the long-wavelength edge of the ytterbium absorption line after 15 min of irradiation with a titanium–sapphire laser. The change in the temperature of a sample was measured indirectly from the deviation of a probe beam of a helium–neon laser due to a temperature-induced variation in the refractive index of the glass. Direct temperature measurements using a nitrogen-cooled mid-infrared camera were also performed.

Around the same time, Rumbles and Clark, of the Imperial College of Science, Technology, and Medicine, London, have implemented the laser cooling of a liquid solution of Rhodamine 101 in acid-containing ethanol by exciting Rhodamine molecules into the long-wavelength edge of the absorption line with argon-laser radiation during 4 h [7]. An indirect procedure of temperature measurements based on temperature-dependent variations in the intensity of fluorescence excited with a probing helium–neon laser has revealed the lowering of the temperature by 3 K with respect to room temperature.

The authors of both experiments attribute the effect of laser cooling to the so-called anti-Stokes laser cooling. A diagram of this process is shown in Fig. 1. First, pump laser radiation detuned from the resonance transfers electrons from a vibrational sublevel 1' of the ground state to a purely electronic excited level 2. Then, spontaneous emission through direct and anti-Stokes indirect transitions transfers the system to the ground level 1. Direct transitions play the main role in this process [8, 9], although anti-Stokes indirect transitions may also be of crucial importance under certain conditions [10]. Anyway, the process as a whole is called the anti-Stokes process because the frequency of emitted radiation is higher than the frequency of absorbed radiation. Since the energy (frequency) of pump photons is insufficient to give rise to purely electronic transitions, only atoms vibrating with the frequency equal to the difference of freq-

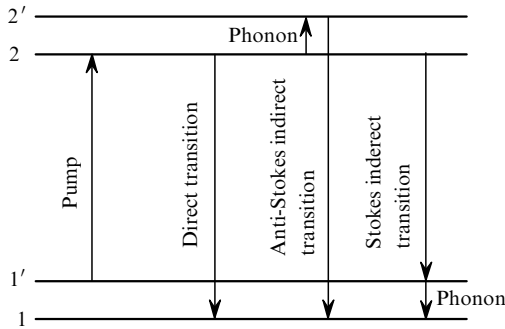
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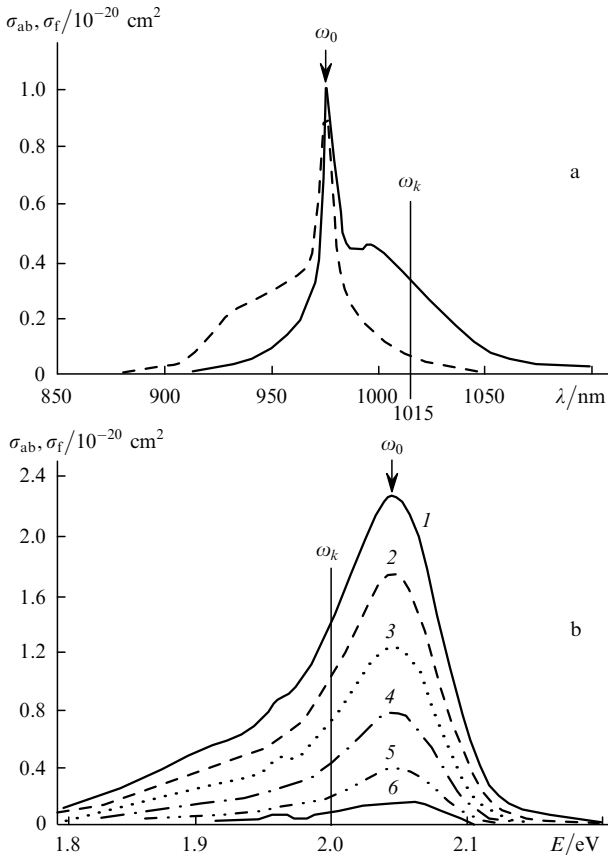
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**Figure 1.** Approximated diagram of energy levels and transitions involved in anti-Stokes laser cooling.

encies of the electronic transition and photons and lying above the electronic ground level 1 may be excited. The energy of atomic vibrations under these conditions is transformed into the excitation energy of the electronic shell. Subsequently, this energy is removed from the sample by photons of spontaneous emission.

Absorption and fluorescence spectra of ytterbium ions in glass and Rhodamine molecules in ethanol are shown in Fig. 2. These spectra demonstrate that the excitation frequency fell within the long-wavelength wing of the absorption line of impurity molecules, which confirms the



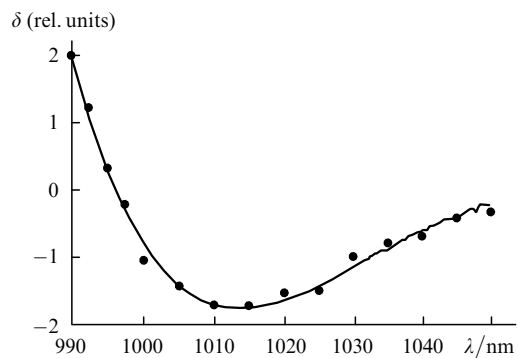
**Figure 2.** Cross sections of absorption lines  $\sigma_{ab}$  (dotted curves) and fluorescence  $\sigma_f$  (solid curves) for ytterbium ions in heavy metal–fluoride glass as functions of the wavelength (a) and for Rhodamine molecules in acid-containing ethanol (b) as functions of the energy for temperatures 310 (1), 290 (2), 270 (3), 250 (4), 220 (5), and 200 K (6) [6, 7].

anti-Stokes model of laser cooling illustrated in Fig. 1. In the case of ytterbium, the highest efficiency of cooling is achieved when the radiation wavelength is detuned by 17 nm from the central wavelength of emission, which corresponds to the excitation from the lower sublevel of the Stark multiplet of ytterbium ions ( $179 \text{ cm}^{-1}$ ) in glass [13]. A weakly pronounced phonon wing allows us to employ the model of weak dipole–phonon coupling in our analysis. In the case of Rhodamine in ethanol, the optimal detuning was equal to 9 nm.

The most impressive progress in the laser cooling of solids was achieved in Los Alamos in experiments with fibre samples due to the high optical purity and a small surface area of these samples. Ytterbium-doped optical fibres allowed laser cooling first by 16 K [14] and then by 21 K [15] to be implemented below room temperature. In what follows, we shall examine these experiments in greater detail.

The first series of experiments was devoted to the laser cooling of bulk samples. Continuous-wave radiation of a titanium–sapphire laser was employed as a pump in these experiments. This radiation was focused on a polished sample. Probe radiation of a helium–neon laser irradiated the same sample, propagating in the opposite direction with a small transverse shift relative to pump radiation. Laser cooling gave rise to a temperature-induced gradient of the refractive index in the sample, resulting in a deflection of probe radiation. This deflection was measured with a detector sensitive to the beam position.

The beam deviation as a function of the pump wavelength is shown in Fig. 3. The maximum cooling, as can be seen from this figure, is achieved for a pump wavelength of 1015 nm, which corresponds to the absorption of radiation due to vibrations related to the lower sublevel in the Stark multiplet of ground-state ytterbium in glass. These results were confirmed by direct measurements of the cooling temperature using an infrared camera, which indicated the lowering of the temperature by 0.3 K.



**Figure 3.** Normalised deviation  $\delta$  of the probe beam as a function of the wavelength [6].

Laser cooling was studied for fibres with outer diameters of 400, 250, and 135  $\mu\text{m}$ . As the fibre diameter decreases from 400 to 250  $\mu\text{m}$ , the efficiency of laser cooling increases. However, for a 135- $\mu\text{m}$ -diameter fibre, the cooling efficiency lowers again, which is apparently due to impurities in the material of the fibre, giving rise to heat release. The most significant cooling (21 K below room temperature) was obtained for a fibre with an outer diameter of 250  $\mu\text{m}$  and an inner diameter of 175  $\mu\text{m}$ . The decrease in the

temperature due to laser cooling in these experiments was measured from the temperature-dependent fluorescence.

Laser-cooling experiments were also performed at temperatures of 150 and 100 K. Cooling in these experiments was indicated by a deviation of a probe beam from a helium–neon laser, while the absolute change in the temperature was not measured. The observed tendency of changes in the efficiency of laser cooling with a temperature variation indicates the possibility of cooling down to cryogenic temperatures. Temperature changes in the cryogenic range can be measured from the temperature-sensitive decay of the photon-echo intensity [16].

### 3. Theory

Anisotropic impurity molecules are involved in vibrational librations with respect to their equilibrium positions in a molecular crystal. Since the direction of their transition electric dipole moment is unambiguously related to molecular symmetry axes, these librations modulate the constant of interaction of a molecule with the electromagnetic field, which gives rise to the so-called indirect transitions, when a phonon is absorbed or emitted simultaneously with a photon. Indirect transitions are manifested as narrow lines appearing against the background of the continuous long-wavelength wing of an absorption line.

In the case of the rare-earth ions discussed above, phonons are absorbed through transitions from sublevels of the Stark multiplet. Only after that, phonons are absorbed due to these sublevels. However, phonon processes are very fast, and we can employ the same model Hamiltonian in this case.

The Hamiltonian of a system of impurity ions or molecules in a crystal interacting with an electromagnetic field in the dipole approximation and with the crystal lattice in the model of weak coupling can be written as

$$H = H_0 + H_1, \quad (1)$$

where

$$H_0 = H_d + H_f + H_p \quad (2)$$

is the main Hamiltonian and

$$H_1 = H_{df} + H_{df}^p + H_{pp} \quad (3)$$

is the perturbation Hamiltonian. Here,

$$H_d = \sum_j \hbar \omega_0 S_j^z \quad (4)$$

is the Hamiltonian of the system of impurity ions or molecules in the model of a two-level atom;  $j$  is the number of an impurity molecule;  $\omega_0$  is the frequency of the electronic transition; and  $S_j^z$  is the operator of the  $z$ -projection of the effective spin  $S = 1/2$ ;

$$H_f = \sum_k \hbar \omega_k a_k^+ a_k \quad (5)$$

is the Hamiltonian of the field of photons;  $k$  is the index specifying three components of the wave vector and polarisation of photons;  $\omega_k$  is the frequency of photons; and  $a_k^+$  and  $a_k$  are the operators of creation and annihilation of photons;

$$H_p = \sum_q \hbar \Omega_q b_q^+ b_q \quad (6)$$

is the Hamiltonian of the field of phonons;  $q$  is the index specifying three components of the wave vector and polarisation of phonons;  $\Omega_q$  is the frequency of phonons; and  $b_q^+$  and  $b_q$  are the operators of creation and annihilation of phonons;

$$H_{df} = \sum_{jk} [g_k \exp(i\mathbf{k}\mathbf{r}_j) S_j^+ a_k + g_k^* \exp(-i\mathbf{k}\mathbf{r}_j) S_j^- a_k^+] \quad (7)$$

is the operator of interaction between atoms and the electromagnetic field through the direct transition in the rotating-wave approximation;  $g_k$  is the coupling constant;  $\mathbf{r}_j$  is the radius vector of an impurity atom; and  $S_j^+$  and  $S_j^-$  are the operators raising and reducing the components of the spin operator in cyclic coordinates;

$$H_{df}^p = \sum_{jkq} \{h_{kq} \exp[i(\mathbf{k} - \mathbf{q})\mathbf{r}_j] S_j^+ a_k (b_{-q} + b_q^+) + h_{kq}^* \exp[-i(\mathbf{k} - \mathbf{q})\mathbf{r}_j] S_j^- a_k^+ (b_q + b_{-q}^+)\} \quad (8)$$

is the operator governing the interaction of atoms with the electromagnetic field and the lattice through indirect transitions via some sublevels against the background of the continuous spectrum of phonons in the rotating-wave approximation and  $h_{kq}$  is the relevant coupling constant; and

$$H_{pp} = V^{-1} \sum_{q_1, q_2, q_3, \tau} \Phi^\tau(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3) b_{q_1}^+ b_{q_2}^+ b_{q_3} + \text{H.c.} \quad (9)$$

is the operator governing phonon–phonon interactions;  $V$  is the volume of a unit cell;  $\Phi^\tau(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3)$  is the coupling constant; and  $\tau$  is an arbitrary vector of a reciprocal lattice.

Thus, we consider a system of  $N$  impurity centres in a solid subject to the action of continuous-wave laser radiation at frequency  $\omega$ , which is resonant to the frequency of the  $1' \rightarrow 2$  transition (see Fig. 1). The population of the level  $1'$  is determined by the phonons of the mode  $q$ . Let us introduce the number of phonons in the  $q$ th mode,

$$n_q = b_q^+ b_q \quad (10)$$

and the collective population difference for the working electronic levels,

$$R_z = 2 \sum_j S_j^z. \quad (11)$$

Then, using the method of nonequilibrium statistical operator [17], we derive the following equations for dynamic variables  $D_m$ :

$$\frac{d\langle D_m \rangle}{dt} = \hbar^{-2} \int_{-\infty}^0 dt e^{\varepsilon t} \langle [H_1(t), [D_m, H_1]] \rangle_q, \quad (12)$$

where the parameter  $\varepsilon$  takes into account the irreversible character of the processes under study (this parameter should be set equal to zero after the integration); the angle brackets with a subscript  $q$  denote the averaging over the quasi-equilibrium statistical operator; and

$$H_1(t) = \exp\left(\frac{i}{\hbar} H_0 t\right) H_1 \exp\left(-\frac{i}{\hbar} H_0 t\right). \quad (13)$$

Applying the method of nonequilibrium statistical operator, we arrive at the following set of equations:

$$\begin{aligned} \frac{\partial \langle n_q \rangle}{\partial t} + \mathbf{v}_q \nabla \langle n_q \rangle &= \left[ \frac{\langle n_q \rangle + 1}{\tau_1^s(q)} - \frac{\langle n_q \rangle}{\tau_1^{as}(q)} + \frac{1}{\tau_s(q)} \right] \\ &\times \frac{\langle R_z \rangle + N}{2} + \left[ \frac{1}{\tau_s(q)} - \frac{1}{\tau_{as}(q)} \right] \langle n_q \rangle \langle R_z \rangle \\ &+ \frac{N - \langle R_z \rangle}{2\tau_{as}(q)} - L(n_q), \end{aligned} \quad (14)$$

$$\begin{aligned} \frac{\partial \langle R_z \rangle}{\partial t} &= -(N + \langle R_z \rangle) \left\{ \frac{1}{\tau_1} + \sum_q \left[ \frac{\langle n_q \rangle + 1}{\tau_1^s(q)} + \frac{\langle n_q \rangle}{\tau_1^{as}(q)} \right. \right. \\ &\left. \left. + \frac{1}{\tau_s(q)} \right] \right\} - 2\langle R_z \rangle \left\{ \frac{1}{\tau_r} + \sum_q \left[ \frac{1}{\tau_s(q)} + \frac{1}{\tau_{as}(q)} \right] \langle n_q \rangle \right\} \\ &+ (N - \langle R_z \rangle) \sum_q \frac{1}{\tau_{as}(q)}, \end{aligned} \quad (15)$$

where

$$\begin{aligned} L(n_q) &= \frac{2\pi}{\hbar^2} \sum_{q_1, q_2, q_3} |\Phi^\tau(q_1, q_2, q_3)|^2 \delta(\omega_{q_1} + \omega_{q_2} - \omega_{q_3}) \\ &\times (\delta_{qq_1} + \delta_{qq_2} - \delta_{qq_3}) [\langle n_{q_3} \rangle (1 + \langle n_{q_1} \rangle) (1 + \langle n_{q_2} \rangle) \\ &- \langle n_{q_1} \rangle \langle n_{q_2} \rangle (1 + \langle n_{q_3} \rangle)] \end{aligned} \quad (16)$$

is the integral of phonon–phonon collisions;  $\mathbf{v}_q$  is the velocity of phonons;  $\omega_{q_1}$ ,  $\omega_{q_2}$ , and  $\omega_{q_3}$  are the phonon frequencies;

$$\frac{1}{\tau_1} = \sum_k \frac{1}{\tau_1(k)} \quad (17)$$

is the inverse time of spontaneous relaxation through  $2 \rightarrow 1$  direct transitions;

$$\frac{1}{\tau_1^s(q)} = \sum_k \frac{1}{\tau_1^s(k, q)} \quad (18)$$

is the inverse time of spontaneous relaxation through  $2 \rightarrow 1' \rightarrow 1$  indirect transitions;

$$\frac{1}{\tau_1^{as}(q)} = \sum_k \frac{1}{\tau_1^{as}(k, q)} \quad (19)$$

is the inverse time of spontaneous relaxation through  $2 \rightarrow 2' \rightarrow 1$  indirect transitions;

$$\frac{1}{\tau_r} = \sum_k \frac{\langle n_k \rangle}{\tau_1(k)} \quad (20)$$

is the inverse time of stimulated  $2 \rightarrow 1$  direct transitions;

$$\frac{1}{\tau_s(q)} = \sum_k \frac{\langle n_k \rangle}{\tau_1^s(k, q)} \quad (21)$$

is the inverse time of stimulated  $2 \rightarrow 1' \rightarrow 1$  indirect transitions; and

$$\frac{1}{\tau_{as}(q)} = \sum_k \frac{\langle n_k \rangle}{\tau_1^{as}(k, q)} \quad (22)$$

is the inverse time of stimulated  $2 \rightarrow 2' \rightarrow 1$  indirect transitions [9].

The set of equations derived above is too complicated. Therefore, some simplifying assumptions will be used. In particular, we assume that deviations from the equilibrium phonon numbers are small,  $\langle n_q \rangle \approx n_q^0$ , where  $n_q^0$  is the equilibrium phonon number for a given temperature of the sample. Then, since  $L(n_q^0) = 0$  [18], the collision integral on the right-hand side of Eqn (14) can be neglected. On the other hand, in the case when cooling of a small sample gives rise to small temperature deviations, the term involving the coordinate derivative on the left-hand side of this equation can be approximated with

$$v_q \frac{\Delta n_q^0}{\Delta x} = \frac{n_q^0 - \bar{n}_q}{\tau_1(q)},$$

where  $\Delta n_q^0 = n_q^0 - \bar{n}_q$ ;  $\tau_1(q) = \Delta x / v_q$ ;  $\bar{n}_q$  is the number of thermostat phonons; and  $\Delta x$  is the size of the sample. This approximation corresponds to a spatially uniform situation and a weakly nonequilibrium phonon system. Then, ignoring anti-Stokes indirect processes, we can apply Eqn (14) to derive

$$\begin{aligned} \frac{d \langle n_q \rangle}{dt} &= \left( \frac{\langle n_q \rangle + 1}{\tau_1^s(q)} + \frac{1}{\tau_s(q)} \right) \frac{\langle R_z \rangle + N}{2} \\ &+ \frac{1}{\tau_s(q)} \langle n_q \rangle \langle R_z \rangle - \frac{\langle n_q \rangle - \bar{n}_q}{\tau_p(q)}. \end{aligned} \quad (23)$$

Similarly, Eqn (15) yields

$$\begin{aligned} \frac{d \langle R_z \rangle}{dt} &= -(N + \langle R_z \rangle) \left\{ \sum_q \left[ \frac{\langle n_q \rangle + 1}{\tau_1^s(q)} + \frac{1}{\tau_s(q)} \right] + \frac{1}{\tau_1} \right\} \\ &- 2\langle R_z \rangle \sum_q \frac{\langle n_q \rangle}{\tau_s(q)}. \end{aligned} \quad (24)$$

We should take into account that the system is irradiated only at the frequency of the Stokes indirect transition. Therefore, ignoring the amplification of spontaneous luminescence of the system, we can assume that  $1/\tau_r = 0$ . Summation over the wave vector of phonons then yields

$$\begin{aligned} \frac{d \langle n \rangle}{dt} &= \frac{N + \langle R_z \rangle}{2} \left[ F_1^s(T) + \frac{1}{T_1^s} + \frac{1}{T_s} \right] \\ &+ \langle R_z \rangle F_s(T) - F_p(T) + F_p(T_0), \end{aligned} \quad (25)$$

$$\begin{aligned} \frac{d \langle R_z \rangle}{dt} &= -(N + \langle R_z \rangle) \left[ F_1^s(T) + \frac{1}{T_1^s} + \frac{1}{T_s} + \frac{1}{\tau_1} \right] \\ &- 2\langle R_z \rangle F_s(T), \end{aligned} \quad (26)$$

where

$$\langle n \rangle = \sum_q \langle n_q \rangle \quad (27)$$

is the total number of phonons;

$$\begin{aligned} \frac{1}{T_1^s} &= \sum_q \frac{1}{\tau_1^s(q)}; \quad \frac{1}{T_s} = \sum_q \frac{1}{\tau_s(q)}; \quad F_1^s(T) = \sum_q \frac{\langle n_q \rangle}{\tau_1^s(q)}; \\ F_s(T) &= \sum_q \frac{\langle n_q \rangle}{\tau_s(q)}; \quad F_p(T) = \sum_q \frac{\langle n_q \rangle}{\tau_p(q)}. \end{aligned} \quad (28)$$

In the stationary regime, we have

$$F_s(T)\{2\tau_1[F_p(T) - F_p(T_0)] + N\} + \tau_1[F_p(T) - F_p(T_0)] \times \left[ F_1^s(T) + \frac{1}{T_1^s} + \frac{1}{T_s} + \frac{1}{\tau_1} \right] = 0, \quad (29)$$

where  $T_0$  is the temperature of the thermostat.

In the approximation of high temperatures,  $\langle n_q \rangle = k_B T / \hbar \Omega_q$ , we arrive at

$$AT^2 + BT - CT_0 = 0, \quad (30)$$

where

$$A = \tau_1 \Phi_p (\Phi_1^s + 2\Phi_s); \quad B = \Phi_s N + C - AT_0; \quad C = \tau_1 \Phi_p \left( \frac{1}{T_1} + \frac{1}{T_s} + \frac{1}{\tau_1} \right); \quad (31)$$

$$\Phi_1^s = k_B \hbar^{-1} \sum_q [\Omega_q \tau_1^s(q)]^{-1}; \quad \Phi_s = k_B \hbar^{-1} \sum_q [\Omega_q \tau_s(q)]^{-1}; \quad \Phi_p = k_B \hbar^{-1} \sum_q [\Omega_q \tau_p(q)]^{-1}; \quad (32)$$

and  $k_B$  is the Boltzmann constant.

The solution to Eqn (30) is written as

$$T = (2A)^{-1} [-B + (B^2 + 4ACT_0)^{1/2}]. \quad (33)$$

Expanding the square root as a power series in  $1/T_0$ , we derive

$$T = T_0 - \frac{\Phi_s N}{\tau_1 \Phi_p (\Phi_1^s + 2\Phi_s)}. \quad (34)$$

Hence, we find that the cooling temperature decreases with the growth in the concentration of impurity centres, the increase in the rate of spontaneous relaxation through the direct  $2 \rightarrow 1$  transition, and the weakening of the coupling with the thermostat. For high pump powers ( $\Phi_s \gg \Phi_1^s$ ), we reach the saturation regime,

$$T = T_0 - \frac{N}{2\tau_1 \Phi_p}, \quad (35)$$

i.e., the cooling temperature becomes independent of the pump power. This expression is more general than the corresponding expression from [9], as it includes summation over all the types of vibrations involved in the process under study. Replacing summation in  $q$  in the expression for  $\Phi_p$  by multiplication by the effective number of vibrations  $M$ , we arrive at

$$T = T_0 - \frac{\hbar \Omega_0}{2k_B} \frac{\tau_p N}{\tau_1 M}, \quad (36)$$

where  $\Omega_0$  is the frequency of the active vibration. Note that Eqn (36) coincides with the corresponding expression from [9]. However, the quantity  $M$  now stands for the total number of all the vibrations involved in the process of laser cooling. Setting  $\Omega_0 = 200 \text{ cm}^{-1}$ ,  $\tau_p \approx \tau_1 \approx 10^{-5} \text{ s}$ , and  $N = 10^{16}$  [13], we find that  $M = 10^{16}$  when the sample is cooled by 21 K with respect to room temperature, as was the case in Los Alamos experiments.

Note that Eqn (29) can be also solved in the approximation of low temperatures. To do this, we should take into consideration that

$$\langle n \rangle = \exp \left( -\frac{\hbar \Omega_0}{k_B T} \right)$$

and perform the relevant Laplace transform. However, specific estimates in this case would require the knowledge of the dependences of all the coefficients on  $q$ .

## 4. Conclusions

Thus, the developed theory of laser cooling of impurity crystals shows that the inclusion of spatial inhomogeneity of phonon states brings us to the Eqn (14) whose right-hand side depends on the coupling of photons with impurity atoms. Simple expressions for the temperature of a sample including phonon dispersion were derived for a spatially homogeneous system. Predictions of this theory were compared with the results of experiments on the laser cooling of optical fibres carried out in Los Alamos. This comparison allowed us to estimate the number of phonon states contributing to the process under study and permitted us to infer that Stark sublevels are involved in the process of anti-Stokes laser cooling at room temperature. The developed theory of the anti-Stokes mechanism of laser cooling of impurity solids allows us to suggest that Zeeman sublevels could be employed at low temperatures instead of Stark sublevels, since the energies of Zeeman sublevels correspond to the energies of phonon states populated at low temperatures.

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