Enhanced Laser Cooling Using Ion-Doped Nanopowders: Engineering and Harvesting Atomic Vibrations

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Abstract

This research focused on maximizing the overall cooling rate to cool solids from room temperature to the cryogenic temperature range by advancing the existing theoretical treatments. At the atomic level, laser cooling is described through the energy transfer mechanisms among photons, electrons, and phonons. The cooling rate is dependent on the interactions and properties of the host atoms, the optically-active dopant, and the coupling between these three carriers [1]. Investigated parameters included the electron-phonon coupling coefficient and the phonon density of states (DOS) using the Debye-Gaussian model, both of which affect the phonon-assisted photon absorption rate, the target phonon energy, and the nonradiative decay rate. To enhance the cooling rate, doped nanopowders are used over bulk since the DOS of nanopowders has broader peaks [2].

Background

The ultimate goal in laser cooling research is to create a refrigeration unit capable of reaching the cryogenic temperature range. Since it lacks moving parts, such a device would have a longer lifetime than other coolers. The largest reported temperature drop has been 70 K from room temperature, and the process has been observed as low as 77 K [3].

Laser Cooling Process

The solid to be cooled is composed of an ionic host doped with an optically-active rare-earth ion. Incident photons have a frequency tuned slightly lower than the resonance transition of the dopant. In order for an electron of the dopant to absorb an incident photon, a phonon from the host must also be absorbed so that the sum of the photon and phonon energies equals the resonance transition. The electron then decays back to the ground state either radiatively, with the emission of a photon, or nonradiatively, with the emission of phonon(s) and a photon. Cooling occurs if the average emitted photon has a greater energy than the incident photon.

Cooling Rate Equation

In Figure 1, the cooling rate equation is given in the unit of watt [1]. The variables which vary with the selection of host include the effective mass of the constituent atoms m_{eff}, the electron-phonon coupling coefficient $\varphi'_{e\text{-}p,O}$, the phonon DOS D_p, the target phonon energy E_p, the Bose-Einstein distribution f_p^o, and the internal quantum efficiency $\eta_{e\text{-}ph}$. Other factors from the equation, which vary with the properties of the optically-active dopant, laser irradiation, and the structure of the sample, include the photon-electron coupling coefficient $\boldsymbol{s}_\alpha \bullet \boldsymbol{\mu}_e$, the frequency $\omega_{ph,i}$ and wavelength $\lambda_{ph,i}$ of the laser photons, the number density of the dopant n_d, the length of the solid L, the speed of light u_{ph}, the incident laser power $Q_{ph,i}$, and the average wavelength of the emitted photons $\lambda_{ph,e}$.

$$\dot{S}_{ph-e-p} = \frac{\pi\hbar}{2\varepsilon_o m_{eff}}(\boldsymbol{s}_\alpha \cdot \boldsymbol{\grave{\imath}}_e)^2 \varphi'^2_{e-p,O} \frac{D_p(E_p) f_p^o(E_p)}{E_p^{\ 3}} \frac{\hbar\, \omega_{ph,i} n_d L}{u_{ph}} Q_{ph,i}\left(1 - \frac{\lambda_{ph,i}}{\lambda_{ph,e}} \eta_{e-ph}\right)$$

Figure 1: Cooling rate equation.

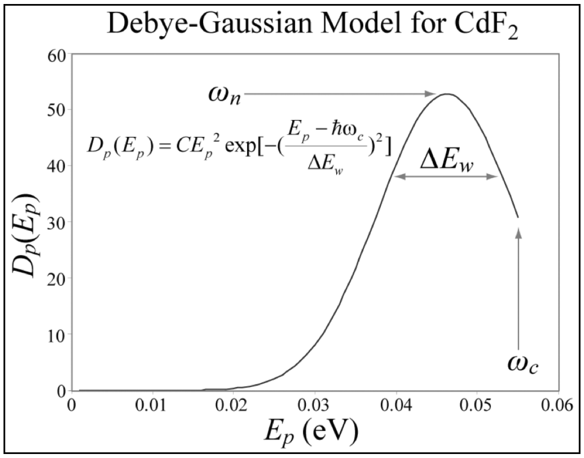

Figure 2: DOS for CdF_2 using the Debye-Gaussian model, shown with the DOS equation, where the constant C is dependent on the cut-off frequency ω_c ($C = 26587.7\ eV^{-3}$ for CdF_2).

Phonon DOS

The density of states (DOS), which gives the number of phonon modes for a given energy, was estimated using the Debye-Gaussian model (Figure 2). The peak value was calculated from the natural frequency ω_n, the cut-off frequency ω_c, and the width of the peak ΔE_w. The area under the curve was normalized to one, so a lower cut-off frequency increased the peak value.

By using the harmonic oscillator model, cut-off frequencies were calculated for various fluorides, chlorides, and oxides from the spring force constant and effective mass between the anion-cation pair. In addition to increasing the peak DOS value, lower cutoffs augment the cooling rate by increasing the internal quantum efficiency and decreasing the target phonon energy. From the calculations, chloride paired with a heavy cation would maximize the cooling rate through these factors.

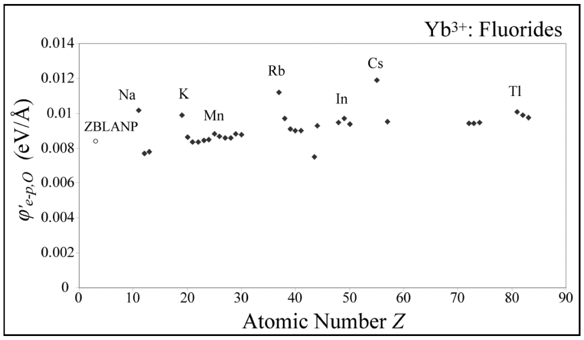

Figure 3: Electron-phonon coupling coefficient for various cations in fluoride glass.

Electron-Phonon Coupling

In the harmonic oscillator model, the dopant absorbs phonons from the vibrations of a neighboring anion. When an anion-cation pair oscillates with respect to the dopant, the anion-dopant bond length changes, changing the potential of the dopant's valence electrons. As the coupling factor is increased through host selection, the same anion-dopant contraction or dilation increases the potential change of the electrons. The electron-phonon coupling coefficient was calculated for Yb^{3+}-doped fluorides (Figure 3). Electron-phonon coupling increases slowly with atomic number and peaks at the alkali metals. Rubidium and cesium fluorides are the optimal cations for maximizing electron-phonon coupling.

Nanopowders

The properties of nanopowders have been found to enhance the cooling rate over bulk materials by increasing the number of energy carriers in laser cooling, specifically phonons and photons [2]. Calculations show that the DOS of nanopowders is on average larger at the relevant phonon energies. Photon scattering in random nanopowders leads to photon localization, where photons do not propagate through the solid but are spatially restricted to a region. This leads to a much higher photon density per input power.

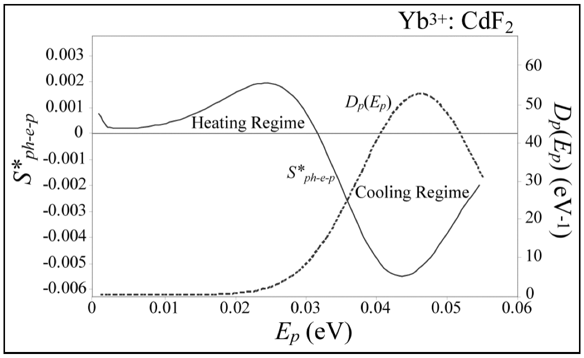

Figure 4: Calculated cooling rate (normalized per unit of input power $Q_{ph,i}$) and normalized DOS vs. target phonon energy.

Conclusions

The selection of an ionic host material has been discussed through the DOS and the electron-phonon coupling coefficient. From the results, ideal cations are rubidium and cesium. Currently, fluoride is used as the anion because it is known to be optically transparent to the laser photons; more research is needed for other anions.

In Figure 4, the normalized cooling rate varies with the target phonon energy. In the heating regime, the target phonon energy is too low, so each instance of radiative decay does not remove a sufficient amount of phonon energy to overcome the heating of nonradiative decay. The target phonon energy which provides the maximum cooling rate (most negative) is shifted slightly to the left of the DOS peak value due to the Bose-Einstein distribution and the E_p^3 term in the denominator of the cooling rate equation.

Acknowledgements

I would like to thank Dr. Massoud Kaviany for allowing me to work in the Heat Transfer Physics Laboratory and Jedo Kim for mentoring me on my project. This project was supported by the National Nanotechnology Infrastructure Network Research Experience for Undergraduates Program and the National Science Foundation.

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