

DEFORMATION COOLING

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A formula is obtained which describes the change in the temperature of a system under adiabatic removal of an external force. As an example, the cooling of a spin system in the ground and excited states due to the removal of elastic stresses is considered. The possibility is considered of using deformation cooling for the polarization of electron and nuclear spins.

THE possibility is considered below of the cooling of a system of interacting particles with orbital and spin magnetism, and with electric quadrupole moments, by means of the change in the symmetry of the intracrystalline fields by the external pressure. As is known from numerous experiments, a phenomenon analogous to the Jahn-Teller effect of free molecules need not necessarily take place in systems which possess translational symmetry. The spin-orbit interaction and the presence of covalent bonds also prevent the appearance of this phenomenon.^[1-7] Therefore, in a number of cases, when the crystal has high symmetry, degeneracy of the energy levels of the system can be maintained in spite of the large intensity of the internal electric field. Here the insignificant change in the symmetry of the crystalline electric field by the pressure can produce a large energy splitting of the degenerate levels.

Let the deformed crystal be in equilibrium at the initial time $t = 0$. If now, after a time t_0 , the deformation is removed without changing the population of the levels, the system of particles is shown to be in a nonequilibrium state ("supercooled") and will not absorb the energy from the other degrees of freedom of the crystal. The adiabatic character of the process of removal of the external influence, which had been imposed on the system, required the satisfaction of the condition

$$\tau_S \ll t_0 \ll \tau_{SL}, \tag{1}$$

where τ_S and τ_{SL} are the characteristic times for establishment of equilibrium within the system and between the system and its surroundings. The finite temperature T_0 of the system, which is obtained as the result of the adiabatic removal of the external force, is calculated from the formula

$$\text{Sp exp} [- (\mathcal{H} + \mathcal{H}_0)/kT] = \text{Sp exp} [- \mathcal{H}/kT_0], \tag{2}$$

where T and $\mathcal{H} + \mathcal{H}_0$ are the temperature and the Hamiltonian of the system at the time $t = 0$, \mathcal{H} is the Hamiltonian of the system at the time $t \gg t_0$, and k is Boltzmann's constant.

When

$$\mathcal{H}_0 = \sum_j^N \mathcal{H}_0^j, \quad \nu_0^{-1} \ll \tau_S, \quad T_0^{-1} \gg kh^{-1}\tau_S,$$

we get

$$T_0 = k^{-1} \left\{ \frac{N^{-1} \text{Sp } \mathcal{H}^2}{2[\text{Sp exp}(-\mathcal{H}_0^j/kT) - R]} \right\}^{1/2} \\ \cong T_S \left\{ 2 \left[\sum_n^R \exp\left(-\frac{E_n}{kT}\right) - R \right] \right\}^{-1/2}, \quad T_S = \frac{h}{k\tau_S}, \tag{3}$$

$\text{Sp } \mathcal{H} = \text{Sp } \mathcal{H}_0 = 0$, $1 \leq n \leq R$, N is the number of particles, $h\nu_0$ is the mean interval of energy in the spectrum of the operator \mathcal{H}_0^j , R is the number of states. For $E_n \ll kT$, we have

$$T_0 = (T_S/T_E) T, \quad T_E = k^{-1} \sum_n^R |E_n| = \frac{h\nu_0 R}{k}. \tag{4}$$

It follows from (2) that, independently of the physical nature of the interactions \mathcal{H} and \mathcal{H}_0 , the adiabatic removal of the external factor \mathcal{H}_0 in the sense of (1) leads to a cooling of the system, described by the Hamiltonian \mathcal{H} , while the multiplicity of the lowering of the temperature is described by the expression (3) or by the coefficient T_S/T_E . The inequality $\nu_0^{-1} \ll \tau_S$ guarantees the adiabatic behavior in the sense of Ehrenfest.

The value of the splitting T_E , brought about by the deformation, can be estimated by the formula

$$T_E = \epsilon G k^{-1} = s P G k^{-1}, \tag{5}$$

where ϵ is the relative deformation, G is the constant of the spin or ion-phonon interaction, s is the elastic modulus, and P is the pressure.

Let us estimate the decrease in the temperature of the spin system of impurity particles situated in

the ground or excited states. Using experimental data, we obtained for $P = 10^9$ dynes/cm², (in °K): $T_E = 1$ for the ground state of Fe²⁺ in MgO;^[2] $T_E = 30$ for the excited state of Eu²⁺ in CaF₂;^[3] $T_E = 10$ for the exchange pairs Cr³⁺ - Cr³⁺ in Al₂O₃^[4] and 1S type of donor levels (As, Sb) in Si;^[5] $T_E = 10^{-6}$ and $T_E = 3 \times 10^{-4}$ for the ¹³³Cs nucleus in a metal^[6] and in ¹¹⁵InSb^[7], respectively. The parameters of the homogeneous broadening are $T_S \sim 10^{-4}$ and $T_S = 10^{-7}$ for electron and nuclear levels, respectively; therefore, $T_0 \sim 10^{-5}$ T and $T_0 \sim 10^{-3}$ T for deformation electron and nuclear cooling. It must be emphasized that, inasmuch as for the excited levels, $T_S T_E^{-1} \ll 1$, $t_0 \sim 10^{-5}$ sec and $\tau_{SL} \sim 10^{-3} - 10^{-4}$ sec, the method of deformation cooling makes it possible to obtain low temperatures in a system of excited particles. It is obvious that the well-known method for adiabatic demagnetization gives no such possibility.

Let us consider, with Eu²⁺ in CaF₂ as an example, the application of excited levels for the polarization of the electron and nuclear spins S which are in this same sample and interact with the Eu²⁺. The first excited level of the Eu²⁺ ion is located at a distance of 24,206 cm⁻¹, has a fourfold degeneracy, and upon uniaxial compression is split into two doublets. For helium temperatures, the inhomogeneous width of the levels is $\sim 1.4^\circ$ K, and $T_S \sim 10^{-4}$ °K is determined essentially by the dipole-dipole interactions, and the lifetime $\tau \sim 10^{-3} - 10^{-4}$ sec. After populating the lower doublet by a monochromatic light pulse, the deformation is removed within a time $\tau_S < t < \tau$, heat flows from the cooled system to the excited Eu²⁺ ions and is then removed from the sample by photons as the Eu²⁺ returns to the ground state ($\tau_S^{-1} \sim 10^5 - 10^7$ sec⁻¹ is the frequency of interaction between the spins S and the Eu²⁺ ions). This makes it possible to obtain in practice the absolute polarization even of electron spins.

In the cases considered, the possible increase in the lattice temperature does not play any role as soon as the condition (1) is satisfied. In principle, cooling of the lattice is also possible if the cooling effect predominates over the heating brought about by the process of removing the deformation. It is evident that for the cooling one must use the deformation reduction of degeneracy and nonmagnetic levels. If the intracrystalline electric field does not possess a center of inversion, then the imposition of the external electric field produces a splitting of the energy levels of the ions.^[8] These splittings are of the same order as the magnetic ones, but the selection rule for the transitions between the excited sublevels are satisfied for the quanta of the thermal vibrations of the lattice, thereby guaranteeing an intense transition of energy from the lattice to the spin system. Therefore, cooling of the crystals by the method of adiabatic removal of the electric field is possible.

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