Observation of a coupled electron-phonon mode in an $LiHoF_4$ crystal

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Slowly decaying (with a time constant on the order of 10^{-2} s) vibrational states with an energy close to 7 cm^{-1} have been observed in experiments on the fluorescent detection of heat pulses in an $LiHoF_4$ crystal. The results are interpreted as being a consequence of the resonance interaction of phonons with the electronic states of Ho^{3+} ions, leading to the formation of a coupled electron-phonon mode.

The motion of phonons with terahertz frequencies in crystals containing rare-earth ions with a rich Stark structure in the ground state is largely determined by the resonance interaction with the electronic states of these ions. This interaction is strongest in "concentrated" crystals, in which the rare-earth ions are elements of a regular structure, and leads to the formation of coupled electron-phonon states. As shown in Ref. 1, the coupled states (coupled modes) are characterized by their own dispersion law and they can propagate with much lower velocities than phonons. The anticrossing of the phonon and electronic branches caused by the formation of coupled states was observed in neutron scattering experiments.[2] There are, however, no published experimental data on energy transfer by electron-phonon modes. The studies of the

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propagation of heat pulses in binary lithium-holmium fluoride ($LiHoF_4$) described in this paper represent the first observation of the motion of strongly coupled electron-phonon modes.

The $LiHoF_4$ crystal has tetragonal symmetry and its physical characteristics are close to those of the well-known system $LiVF_4$. A characteristic of the holmium ion in the crystal used by us is the presence of nearly equal energy gaps between the lowest sublevels of the ground state $^5I_8$ and the luminescing excited state $^5F_5$ (7.7 and 6.7 cm^{-1}, respectively[1)]). This characteristic made it possible to use the technique of fluorescent detection of heat pulses[3] to study the propagation of phonons whose frequencies nearly coincide with frequencies of transitions of ground-state rare-earth ions. The source of nonequilibrium phonons was a thin constantan film which was deposited on the surface of the crystal and heated by electric-current pulses. The duration of the pulses was 3 μs and the peak power was 20 W/mm^2. The phonons were detected by the increase in the intensity of luminescence from the upper Γ_{34} sublevel (symmetry according to Ref. 4) of the $^5F_5$ state. The luminescence was excited by a cw He–Ne laser (632.8 nm, power 1–10 mW). A DFS-24 spectrometer was used to record the luminescence. All experiments were performed at a temperature of 2 K.

As the results of the measurements showed, when an electric pulse is introduced into the heater, pulsed rise of luminescence from the Γ_{34} ($^5F_5$) level is observed in the crystal (in close proximity to the heater as well as at a distance on the order of 1 mm). The recorded pulse has a duration of 10^{-2} s, which is almost three orders of magnitude longer than the typical times required for phonons to propagate over distances of several millimeters in crystals.[5] The difference from the radiative relaxation time of the $^5F_5$ level (4×10^{-5} s; determined optically) is almost as large. The measured time is not equal to the time required for the crystal to cool down as a whole, since the pulse is not observed at distances exceeding 1.5 mm (the sample dimensions were $2\times5\times8$ mm).

To clarify the role of the resonance with transitions in the ground state, we have studied the change in the shape of the recorded pulse in a magnetic field oriented along the fourth-order axis. The experiment was based on the difference in the values of the g-factors of the sublevels of the ground and excited states (see the inset in Fig. 1). This

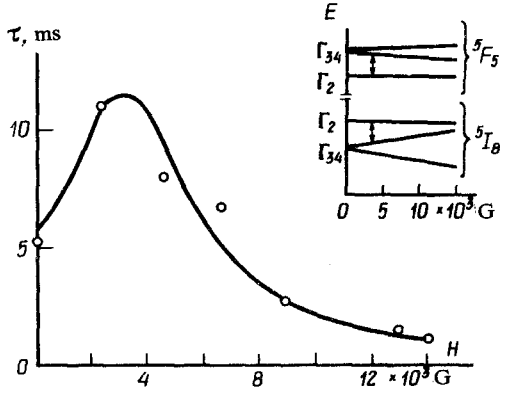

FIG. 1. Field-dependence of the decay time of the recorded pulse. The inset illustrates the splitting of the sublevels of the states $^5F_5$ and $^5I_8$ in a magnetic field.

allowed us to sweep with a magnetic field the gap between the Stark sublevels of the ground state relative to the phonon-detector frequency and thereby to sweep through the region of exact resonance. As is evident from Fig. 1, which shows the results of the measurements, the recorded pulse length, which depends nonmonotonically on the magnetic-field intensity H, has maximum values in fields corresponding to the resonance between the splittings of the ground and excited states. If the spectroscopically measured values of the g-factors are taken into account, the field-dependence of the decay time of the pulse can be described well by a spectral Lorentzian function, which has a half-width of 2 cm^{-1} and which is centered at the resonance frequency. The resonance nature of the change in the duration of the detected pulses shows that the experimentally observed long decay times of the vibrational excitations are a consequence of the existence of coupled electron-phonon modes in the crystal. The width of the resonance is apparently determined by the spectral region of the coupled states, whereas its long duration is explained by the slight contribution of the phonon component, which is responsible for the main dissipative processes—spatial transfer and spectral transformation of energy.

Because of their long decay time, the coupled modes are an effective reservoir for the accumulation of vibrational energy. In the absence of an external heater the exciting light could be a source of excess energy, since the pumping of the $LiHoF_4$ crystal by an He–Ne laser, which is in resonance with one of the upper Stark sublevels of the $^5F_5$ state, causes about 10% of the energy of absorbed photons to be converted into phonons due to nonradiative relaxation along sublevels of the excited and ground states of the holmium ion. As in experiments with heat pulses, the excitation of coupled modes due to absorption of light energy was detected by the luminescence from the upper Γ_{34} sublevel of the $^5F_5$ state. It has been found that for power densities of the continuous optical excitation of $\geqslant 0.1$ W/cm^2 the intensity of luminescence (I_2) from the Γ_{34} ($^5F_5$) sublevel exceeded the values corresponding to its equilibrium temperature population density. The deviation from equilibrium increased with increasing excitation density. The measurements of the magnetic-field dependence of I_2, shown in Fig. 2, are a convincing evidence of the fact that the population in excess of the equilibrium

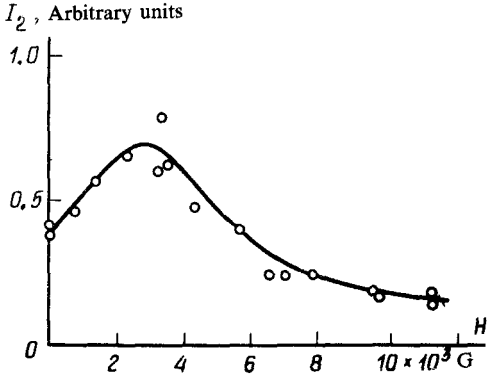

FIG. 2. Field-dependence of the intensity of luminescence from the Γ_{34} ($^5F_5$) level with an optical excitation density of 1 W/cm^2.

population is due to the resonance with the splitting in the ground state. As is evident from a comparison of Figs. 1 and 2, the maximum value of the quantity I_2 occurs in a field in which the resonance condition is satisfied. For excitation densities below 0.1 W/cm^2, the value of I_2 corresponds to the equilibrium population density of the sublevel Γ_{34} ($^5F_5$) at a temperature of 2 K and is independent of the magnetic field.

Based on the data obtained by us we conclude that we have observed in the experiments on the fluorescent detection of heat pulses in the $LiHoF_4$ crystal the propagation of coupled electron-phonon modes with energies on the order of 7 cm^{-1}. We have measured their lifetimes and we estimated the spectral region of their existence. The properties of the coupled state, such as the velocity and anisotropy of propagation, the nature of the coherence, and so on, will be analyzed in greater detail in subsequent studies.

[1)]The gap sizes were determined from the spectroscopic data.

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