LONG-WAVELENGTH OPTICAL PHONONS IN Cd$_{1-x}$Mn$_x$Se AND Cd$_{1-x}$Fe$_x$Se MIXED CRYSTALS

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The results of FIR reflectivity measurements for CdMnSe and CdFeSe are presented. These results are described theoretically using Dynamic Dielectric Function in which two phonon modes are included. The composition dependencies of mode parameters and dielectric constant are discussed.

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The ternary compounds Cd$_{1-x}$Mn$_x$Se and Cd$_{1-x}$Fe$_x$Se belong to the wide class of Diluted Magnetic Semiconductors (DMS) which properties were strongly investigated in the last decade [1]. In these mixed crystals the light magnetic ion substitutes the heavy cadmium one in the wurtzite type lattice. The Raman and preliminary FIR data [2, 3] show the two-mode behavior of CdMnSe crystals. The goal of the present paper is to extend the knowledge about the long-wavelength optical phonons and dielectric properties of this material.

The crystals have been grown by modified Bridgman method in the Institute of Physics of the Polish Academy of Sciences. All given here compositions are technological. In the case of CdMnSe they are in the range from 0.0 up to 0.30 and in the case of CdFeSe they are 0.03 and 0.10. Samples are cut with c-axis parallel to the surface. The reflectivity measurements have been done at RT with unpolarized light using vacuum grating and Fourier spectrometers.

For the both materials CdMnSe and CdFeSe in the reflectivity curves two maxima are clearly seen. Both are composition dependent. Low energy one is due to the host lattice (CdSe) reststrahlen band. The additional structure is connected with Mn local mode evolving to the hypothetical wurtzite type MnSe optical phonons.
The existence of the additional structure in reflectivity and its dependence of composition proofs that these materials exhibit two mode behavior, what has been more detailed discussed in [3].

In such a case the optical properties of the material could be described by the following Dynamic Dielectric Function (DDF)

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{F_{\text{TO}} \omega_{\text{TO}}^2}{\omega_{\text{TO}}^2 - \omega^2 + i\Gamma_{\text{TO}} \omega} + \frac{F_{\text{LT}} \omega_{\text{LT}}^2}{\omega_{\text{LT}}^2 - \omega^2 + i\Gamma_{\text{LT}} \omega},$$

where $\varepsilon_\infty$ is the high frequency dielectric constant, $F_j$ is the oscillator strength of the transverse mode (host crystal for $j = \text{TO}$ and impurity local mode for $j = \text{LT}$), $\omega_j$ and $\Gamma_j$ are the frequency and dumping of the $j$-mode, respectively.

The experimental reflectivity curves were fitted with theoretical one described by the mentioned DDF. The parameters established in this way are discussed further.

In Fig. 1a we present the composition dependence of $\varepsilon_\infty$. As it is clearly seen within the accuracy of 15\% (scatter of experimental data) this parameter is composition independent what agrees with the estimations made for CdMnTe in [6, 7].

If one can treat the local mode and host lattice mode as independent oscillation of two (namely CdSe and MnSe) sublattices then the ratio of the oscillator strengths of these modes $F_{\text{TO}}/F_{\text{LT}}$ should be proportional to the function $x/(1-x)$. As it is seen from Fig. 1b, where the curve is drawn with proportionality factor 0.7, it is the case. The value of the factor suggests that the oscillator strength of TO mode in hypothetical MnSe is about 30\% smaller than this in CdSe. In the case of iron compound this value could be smaller of about 50\%.
In Fig. 2 we compare our data of composition dependence of the mode energies with results calculated in [2] on the basis of Modified Random Element Isodisplacement model [4]. A systematic shift for only local mode energies is seen. This discrepancy could be due to the temperature differences because calculations made in [2] were based on unpublished Raman scattering data obtained at LHeT. It should be pointed out that our data give information about both: longitudinal and transverse modes, when these presented in [2] were for longitudinal modes, only. Further detailed investigations for different temperatures and with polarized light are needed to full explanation of mentioned discrepancies.

We have found that the energies of the local mode for Mn and Fe are within experimental error the same. Because the masses of these two atoms are almost the same, it means that the force constants between Mn–Se and Fe–Se are the same in spite of the differences in the electronic structure of these two magnetic atoms. It should be stressed that the oscillator strength of the Fe local modes is much weaker than this for Mn. And this is probably due to the different electronic structure of ions in the host lattice and hence different coupling of electronic states with the lattice vibrations.

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References