Magnetic Stark effect in layered GaSe crystal

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The magnetic Stark effect is used, for the first time ever, to measure directly the translational mass of excitons in layered GaSe. It is shown that despite the clearly pronounced layered structure of the crystal, the carrier motion in GaSe is practically isotropic.

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The layered structure of crystals of the III–VI group leads to a strong anisotropy of their mechanical effects. It is of interest to assess the extent to which it is accompanied by anisotropy of the electronic states. It was assumed that because of the weak interaction between the crystal layers, the motion of electrons and holes in such systems is essentially confined to the interior of the layer, i.e., is almost two-dimensional, and this should, in particular, manifest itself in the exciton spectra.[1,2]

The most important characteristic that determines the real character of the motion of the electrons and holes in layered crystals is the effective mass of the carriers or excitons in a direction perpendicular to the plane of the crystal layers. The translational mass of the exciton manifests itself most strongly in the presence of the so-called magnetic Stark effect on free excitons.[3] When an exciton moves in a magnetic field, it is acted upon by a force equivalent to the action of an electric field

$$\mathbf{F}_L = \frac{1}{c}[\mathbf{v}\times\mathbf{H}] = \frac{1}{c}\left[\frac{\hbar\mathbf{K}}{M}\times\mathbf{H}\right], \qquad (1)$$

where $\mathbf{v}$ and $\mathbf{K}$ are respectively the velocity and wave vector of the exciton, and M is the translational mass of the exciton. The shifts of the exciton levels in a magnetic field are determined (if spin is disregarded) by the diamagnetic level shifts, by the orbital Zeeman effect, and by the additional splitting of the excitonic states in the field $\mathbf{F}_L$. By applying an external electric field $\mathbf{F}$ it is possible either to enhance or cancel out the contribution to the level splitting from the action of the field $\mathbf{F}_L$. Comparing the pattern of the splitting of the exciton levels at different mutual orientations of the fields $\mathbf{F}$ and $\mathbf{F}_L$ we can directly determine the value of the field $\mathbf{F}_L$ and consequently the effective mass of the exciton.

In the present paper we investigate the Stark effect and the magnetic Stark effect for a free exciton $n=2$ in a layered crystal GaSe.

The electric field was applied with the aid of silver electrodes that produce almost ohmic contacts. To obtain magnetic fields, we used a superconducting solenoid. The incident light propagated parallel to the optical axis C of the crystal, perpendicular to the plane of the crystal layers. The electric and magnetic fields were oriented perpen-

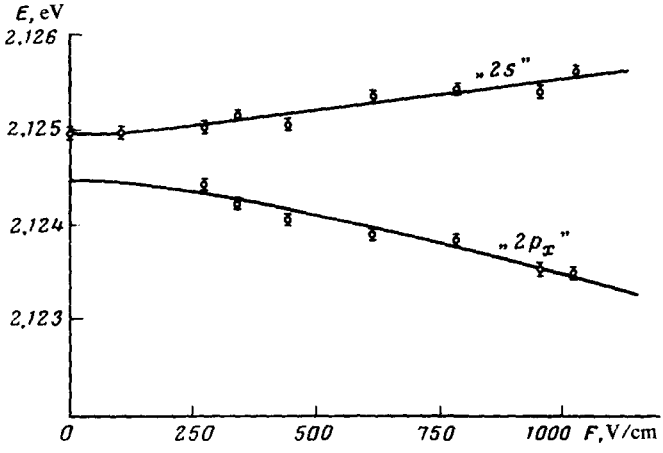

FIG. 1. Stark effect on the state $n=2$ of an exciton in GaSe. Circles–experimental values, solid line–theory.

dicular to the propagation direction of the light, the z axis, along the axes x and y, respectively. The crystals were immersed in liquid helium at $T=2$ K. The crystal transmission spectra were photographed with a diffraction spectrograph having a dispersion 6.5 Å/mm. In the absence of the fields, the exciton absorption lines $n=1$, 2, and 3 were distinctly observed.

Stark effect. In an electric field, the $n=2$ line splits into a doublet (Fig. 1). The intensity of the long-wave component increases with increasing electric field intensity F. The short-wave component of the doublet is due to transitions to the $2s$ state of the exciton, while the long-wave component is due to transitions to the $2p_x$ state. The flareup of the long-wave component is due to the mixing with the allowed $2s$ state. Extrapolation of the experimental results to zero field yields for the initial splitting (due to the anisotropy of the crystal of the $2s$ and $2p_x$ states of the exciton in GaSe a value $\Delta=0.4$–0.6 meV, in full agreement with the results of magneto-optical investigations.[4]

Magnetic Stark effect. In a magnetic field, the $n=2$ line is also split into a doublet.

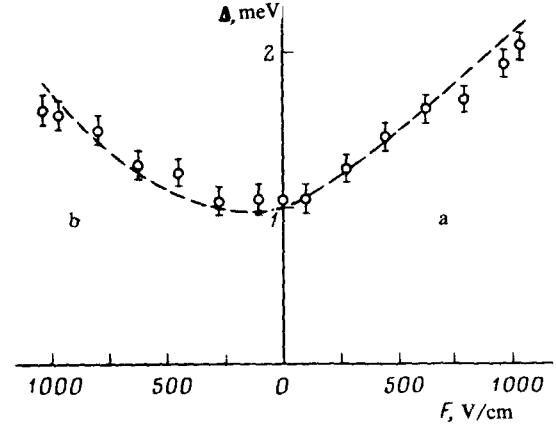

FIG. 2. Splitting of the line $n=2$ in the magnetic Stark effect in GaSe. The direction of F is fixed, $H=32$ KOe=const. a–$[\mathbf{v}\times\mathbf{H}]\mathbf{F}>0, H=H_y$; b–$[\mathbf{v}\times\mathbf{H}]\mathbf{F}<0, H=-H_y$. Circles–experimental values, dashed–curves calculated by perturbation theory.

The doublet components, as shown by a theoretical analysis,[4] are of the same origin as the corresponding components of the doublet in the Stark effect, i.e., they stem from the $2s$ and $2p_x$ states [1]. Application of an electric field leads to an additional shift of the doublet components. The splitting pattern is not symmetrical with respect to reversal of the direction of the magnetic field, i.e., at a given value of $\mathbf{F}$ it depends on the sign of $\mathbf{H}$ (Fig. 2). This phenomenon is the result of the existence of the field $\mathbf{F}_L$. In fact, the splitting of the components depends, in particular, on the total electric field $\mathbf{F}+\mathbf{F}_L$ acting on the exciton. Reversal of the sign of $\mathbf{H}$ changes the sign of the field $\mathbf{F}_L$, and consequently also the summary electric field. Therefore the splitting of the components at fixed $\mathbf{F}$ should depend on the sign of the magnetic field, i.e., it should be asymmetrical with respect to reversal of $\mathbf{H}$, and should be larger at $\mathbf{F}\uparrow\uparrow\mathbf{F}_L$, i.e., at $[\mathbf{v}\times\mathbf{H}]\cdot\mathbf{F}>0$. Both effects were indeed observed in experiment (Fig. 2). The splitting reaches a maximum at $\mathbf{F}+\mathbf{F}_L=0$, i.e., at $\mathbf{F}=-\mathbf{F}_L$. Thus, the position of the minimum of the splitting yields directly the value of the field $\mathbf{F}_L$, from which we can determine the translational mass of the exciton. An analysis of the experimental plot on Fig. 2 yields $\mathbf{F}_L=170\pm30$ V/cm. Using this value of the field and the value of the wave vector of the light-induced excitons $K=2\pi n/\lambda=3.2\times10^5$ cm^{-1}, we obtain from (1) the translational mass of the exciton across the layers $M=(0.7\pm0.1)m_0$. Using this value of M and the experimentally known values of the diamagnetic-shift parameter $\sigma=4.3\times10^{-5}$ meV/kOe^2, of the reduced exciton mass in the plane of the layer $\mu_x=0.14m_0$, and of the ratio of the reduced masses $\mu_x/\mu_z=1.2$,[4] we can calculate the theoretical dependence of the splitting of the $n=2$ line in the magnetic Stark effect in GaSe (Fig. 2). From the condition of best agreement between theory and experiment, we can obtain the effective masses of the carriers bound into the exciton. Their masses turn out to be almost isotropic: $m_x^e=(0.18\pm0.01)m_0$, $m_z^e=(0.15\pm0.01)m_0$, $m_x^h=(0.61\mp0.14)m_0$, $m_z^h=(0.55\pm0.10)m_0$. The use of the obtained parameters makes it also possible to explain without contradiction the Zeeman effect in the GaSe crystal.

Thus observation of the magnetic Stark effect in GaSe crystals points clearly to the presence of exciton motion across the crystal layers, and the small value of the translational mass in this direction, smaller than m_0, shows that the motion in this direction is, unexpectedly, relatively easy. Consequently, the clearly pronounced anisotropy of the mechanical and crystallographic properties of layered crystals is still not a sufficient condition for the manifestation of a similar anisotropy in the properties of any other energy band separately considered, and can be quite readily accompanied by the presence of three-dimensional almost-isotropic energy bands in the crystals.

[1]The spin splitting is small and was not observed in fields $H\lesssim30$ kOe.

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