Optical alignment of excitons

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Alignment of the excitons in a GaSe crystal was observed following excitation with linearly polarized light. The effect of a magnetic field on a system of aligned excitons was observed for the first time. The effective lifetimes of the exciton in the ground and excited states were measured.

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An investigation of the polarized luminescence due to optical orientation or to alignment of atoms is one of the most fruitful trends of modern atomic spectroscopy. Measurement of the degree of polarization of luminescence when different external factors influence the atoms, primarily magnetic fields (the Hanle effect), makes it possible in this case to obtain important information on the atomic constants. The excitons in the crystals are "quasiatoms." They are therefore characterized to a considerable degree by the same phenomena as atoms in gases. One can expect the excitons not only to become oriented, [1,2] as are the free carriers, [3] following excitation by circularly polarized light, but also to become aligned when excited by linearly polarized light. This effect was predicted theoretically in[4]. In this study we have detected alignment of the excitons in a GaSe crystal and observed for the first time the Hanle effect on aligned excitons.

The exciton luminescence was excited in the region of excited states of the exciton with $n \geq 2$ by an optical band of spectral width of approximately 0.03 eV with a maximum radiation 2.14 eV from a DRSh-100 ultrahigh pressure mercury lamp. The exciting light was incident normally on the surface of the crystal, parallel to its optical C axis, and the radiation was registered at a small angle to the propagation direction of the exciting light. If the propagation direction of the exciting light is taken to be the Z axis, and the direction of the oscillations of the electric vector in the incident light wave to be the Y axis (which remains unchanged during the experiment), then the degree of polarization of the radiation is

$$P = (I_y - I_x) / (I_y + I_x), \qquad (1)$$

where I_y and I_x are the intensities of the radiation polarized in directions parallel and perpendicular to the polarization of the exciting light, respectively. We registered the radiation produced upon annihilation of the exciton Γ_6 with $h\nu = 2.1105$ eV in a state $n = 1$. The transitions to this state are allowed only for $E \perp C$ polarization of the light. The excitons in the state Γ_6 are characterized by a spin projection $M_z = \pm 1$ on the C axis, [5] and linearly polarized light excites a coherent superposition of these states.

Figure 1 shows the exciton emission spectrum for two different polarization states (relative to the exciting light). It is seen that the emission of the free exciton Γ_6 (2.1105 eV) is polarized in the same plane as the exciting light, with a high degree of polarization $P = 0.88 \pm 0.05$. The conservation of the linear polarization of the exciton radiation when excited by linearly polarized light indicates an alignment in the exciton system. We note that the linear polarization of the excitation is not conserved for the luminescence of bound excitons with $h\nu_a = 2.097$ eV and $h\nu_b = 2.089$ eV[6] under the same conditions. According to[4], this effect is expected for excitons that are bound with neutral centers.

EFFECT OF LONGITUDINAL MAGNETIC FIELD (HANLE EFFECT)

In a longitudinal magnetic field we observed depolarization of the radiation. This effect is due to the disruption of the initial coherence of the states of the electron with $M_z = \pm 1$ by the magnetic field. The form of the $P(H)$ signal is shown in Fig. 2 (curve 1). We see that $P(H)$ decreases rapidly with increasing H, goes through zero, and reverses sign. A similar behavior of the signal in the Hanle effect is observed in atomic spectroscopy following cascade excitation of luminescence. In the simplest case, when there are three levels, a ground level (0) and two excited levels (1 and 2), with the excitation carried out at the upper level and the luminescence observed for the $1 \rightarrow 0$ transition, the

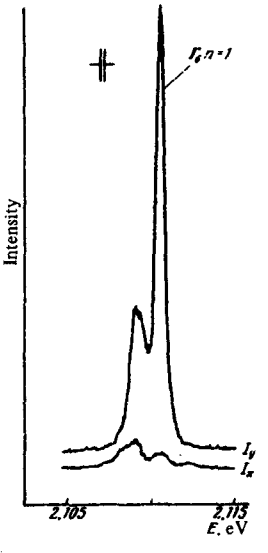

FIG. 1. Exciton-luminescence spectra of GaSe crystal in two different polarizations I_y and I_x, $T = 1.6$ °K.

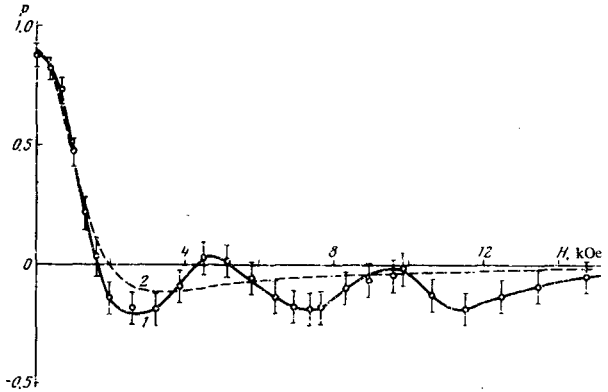

FIG. 2. Plot of the degree of polarization of the exciton luminescence $P(H)$ at $T=1.6$°K. Curve 1—experiment, 2—calculated plot.

degree of polarization is[7]

$$P \sim \frac{\alpha(\alpha - x^2)}{(1+x^2)(\alpha^2+x^2)}, \qquad (2)$$

where $\alpha=(\Delta H)_1/(\Delta H)_2$, $x=H/(\Delta H)_2$, and $(\Delta H)_i$ is the half-width of the Hanle signal in resonant excitation, $(\Delta H)_i=2mc/\tau_i e g_i$, where g_i is the g-factor and $e\hbar g_i H/2mc=\hbar\omega_i$ is the Zeeman splitting of the i-th level.

Under the conditions of our experiment, we undoubtedly have cascade excitation of the exciton state $n=1$, principally via the exciton state $n=2$. This is also evidenced by the high degree of polarization of the exciton luminescence $P(0)=0.88$, since the onset of linearly polarization radiation of excitons is impossible when they are produced from free carriers.[8] This is confirmed by the fact that there was no linear polarization of the exciton line $n=1$ following excitation of the exciton luminescence by the 546-nm mercury line with quantum energy $h\nu=2.27$ eV, which is higher than $E_g=2.130$ eV. In our case, it is logical to set in correspondence the level 0 (the ground state of the crystal) with the level 1 (exciton in state $n=1$) and level 2 (exciton in state $n=2$). Curve 2 of Fig. 2 corresponds to the calculated plot of (2) at $g_{\parallel 1}=g_{\parallel 2}=2.7$.[5] The good agreement between the calculated and experimental curves in the region of the principal maximum of $P(H)$ (near $H=0$) gives for the lifetimes the value $\tau_1=\tau_2=2\times10^{-11}$ sec. Thus, from the waveform of the signal obtained in the Hanle effect under cascade excitation we can obtain the characteristics of both excited exciton levels. Some deviation of the experimental values from the relation (2) can be due, first, to the fact that no account was taken in (2) of the influence of the higher excited states of the exciton with $n\geq 3$, and second, because the transition from $n=2$ to $n=1$ in crystals proceeds principally not optically but with participation of phonons; this, however, should not greatly alter the form of $P(H)$. The presence of two additional maxima of $P(H)$ at $H=4.7$ and 9.6 kOe can be due to the crossing of the fine-structure components of the exciton levels in the magnetic field.

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