Rotation of Linearly Oriented Polaritons in a Magnetic Field

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Under magnetic field in the Faraday configuration, linearly oriented excitonic polaritons are subject to a kind of "resonant Faraday effect." It is first reported here, in the case of CdSe. An energy-dependent lifetime for excitonic polaritons from the bottleneck region can be deduced and is found to be at maximum 7×10^{-12} sec.

It is now well established that optical orientation of excitons in semiconductors can be achieved by linearly polarized light.[1-3] This effect, reported first for CdS,[1,3] is characteristic of a conversion of exciting photons into luminescence photons which only involves free exciton states. Such a process is due to LO-phonon-assisted "indirect" absorption, which creates straightforwardly excitons in their bands; then these excitons decay rapidly to the bottom of the band by LO-phonon cascade. In impure samples, this process of exciton creation is predominant over the ordinary band-to-band absorption followed by exciton formation from electron-hole pairing. This is related to the fact that free electrons and holes decay nonradiatively by trapping on ionized defects, instead of forming excitons.[4]

Such samples are also characterized by short lifetimes and, consequently, incomplete thermalization of excitons. These conditions are favorable to the optical orientation of these excitons. For light propagating parallel to the c axis of a wurtzite-type crystal, an eigenstate of light polarization (belonging to a twofold degenerate subspace) can be converted through the absorption process into an eigenstate of the crystal belonging to the Γ_5 twofold degenerate subspace. Thus, for linearly polarized light, linear orientation of excitons is achieved and the angular momentum transfered to the crystal is transverse. Then, in a magnetic field parallel to the c axis, one can observe the analog of the Hanle effect for circular orientation (nonoccuring for Γ_5 excitons in wurzite-type material because of their zero transverse gyromagnetic factor). The longitudinal magnetic field dephases the two components Γ_{5+} and Γ_{5-} of the linear exciton. If one neglects the interaction with other excitonic levels (such as the Γ_6 level), and introduces two phenomenological parameters τ (lifetime) and T_2 (transverse disorientation time),[5] the result is a rotation α of the mean polarization plane (with regard to the exciting laser polarization) of the emerging photons, and a lower degree of linear polarization P referred to the properly rotated axis.

If $\hbar\omega = g_{ex}^*\mu_B H$ is the energy splitting of the two states Γ_{5+} and Γ_{5-} induced by the magnetic field H, if T_2' is the polarization lifetime defined by

$$T_2'^{-1} = T_2^{-1} + \tau^{-1},$$

and if P_0 is the degree of polarization in zero magnetic field, then we get

$$\tan 2\alpha = \omega T_2', \qquad P = P_0(1+\omega^2 T_2'^2)^{-1/2} = P_0 \cos 2\alpha . \tag{1}$$

The first-order effect in magnetic field intensity is the rotation of the plane of polarization, and can be fruitfully understood in terms of a "resonant Faraday effect." This effect has been predicted by Bir and Pikus,[6] and by ourselves with inclusion of a report of a tentative experiment in CdS[1]; it gave negative results because of experimental limitations. We wish to present here the first observation of this effect.

The experiment is performed in CdSe because of the availability of good signals with the use of a cw tunable cresyl-violet-perchlorate dye laser. The samples are cleaved from larger single crystals grown from the vapor phase. Because of the geometry of the experiment, the surface is cleaved perpendicular to the c axis. This requirement excludes the use of platelets. The luminescence is dispersed by a grating monochromator (18 Å/mm), followed by a photomultiplier and demodulated by a lock-in system. The samples, mounted without strain, are immersed in pumped liquid helium ($T\sim 2$°K). An optical superconducting coil, in the Faraday configuration, provides up to 60 kG. The degree of optical orientation and the rotation of the polarization plane are measured using a rotating-polarizer method. The best samples tested must doubtless be considered as samples of the "impure type" described above, but still good enough for the free exciton-polariton luminescence to be observed for any exciting wavelength.

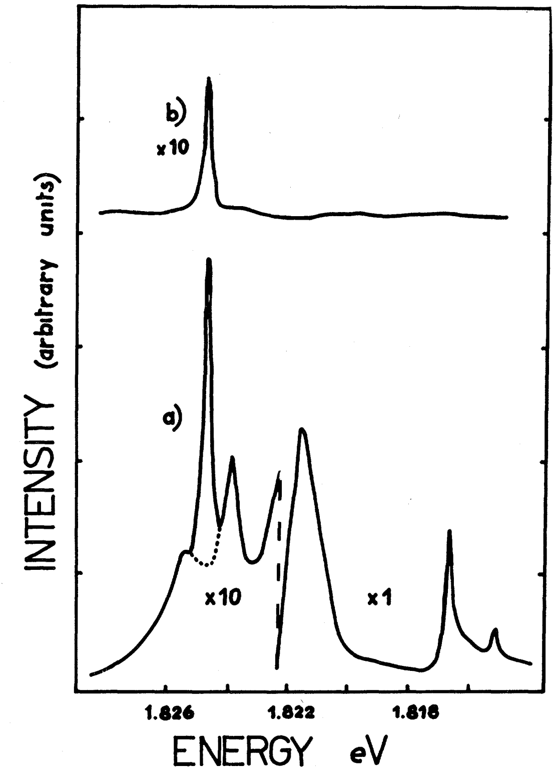

FIG. 1. (a) Fluorescence spectrum ($\sigma^x + \sigma^y$) for exciting energy situated at 1.8975 eV that is two times the LO-phonon energy (2× 26.4 meV) above the resonant peak (1.8248 eV). The permanent spectrum, independent of exciting energy, is shown by a dashed line. (b) Polarization spectrum ($\sigma^x - \sigma^y$) in the same conditions and with the same scale. The signal is zero out of the resonant peak. Thus only this peak is oriented, with a degree of polarization $(\sigma^x - \sigma^y)/(\sigma^x - \sigma^y)$ of +42%.

The luminescence spectra in the excitonic region can be divided into two parts [see Fig. 1(a)]: (i) There is a "permanent luminescence," which is rather independent of the exciting energy. It includes, mainly, the free-exciton-polariton line (1.8254 eV, 0.5 meV wide); a small peak (1.8239 eV, 0.3 meV wide), which may be associated with excitons bound to ionized donors[7]; and the I_2 line (1.8216 eV, 1 meV wide) and the I_1 line (1.8167 eV, less than 0.16 meV wide), which correspond respectively to excitons bound to neutral donors and acceptors. (ii) Superimposed over this "permanent" spectrum, one can observe up to three LO-phonon replicas of the exciting energy as rather sharp peaks (however, wider than the laser linewidth). They are interpreted as luminescence from nonequilibrium excitons created by the indirect absorption described above. From a correct and global point of view, they are the emergence of polaritons scattered from the initial state by some LO and acoustical phonons.[4,8] The magnitude of these peaks (or Raman-like peaks) is very dependent on the number of LO phonons involved and on their energy. They are comparable, or even greater, in magnitude to the permanent polariton line when they are situated around the polariton bottleneck[9] energy and correspond to a two LO-phonon replica. Thus, in this case which we will essentially deal with hereafter, the exciting photons create, by two LO-phonon successive emissions, nonequilibrium polaritons in the bottleneck region.

The polarization spectrum [Fig. 1(b)], i.e., the difference between the intensities of luminescence polarized parallel and perpendicular to the exciting laser polarization, stresses the distinction to be made between the permanent spectrum and the resonant peak: (i) The I_2 and I_1 lines, as well as the "permanent" polariton line (which results from the emergence of polaritons created by electron-hole pairing), are not linearly polarized; this fact can be justified theoretically.[3] (ii) In contrast, the Raman-like peaks are strongly polarized, the degree of polarization lying between 40 and 80%, depending on their energy position.

As will be shown in a forthcoming paper,[10] the polarization spectra, in linear as well as in circular polarization (which we do not discuss here), corroborate the separation of the luminescence spectra into permanent and resonant contributions.

The results of magnetic field measurements are summarized in Fig. 2 for one exciting energy where the effect is important. The theoretical predictions are well verified for a field smaller than 50 kG. The detailed spectral dependence of these effects and their relation to the degree of polarization will be published elsewhere.[10] The important facts are that the rotation α falls to zero outside the bottleneck energy region. Since we have calculated that the exciton g^* factor does not change significantly in the polariton region of interest (it is taken to be[7] 0.69), the variation of the angle α with energy is due to the variation of T_2'. The value of T_2' is calculated from the slope of $\alpha(H)$ at a fixed energy. Its maximum value is found to be $(7 \pm 1)\times 10^{-12}$ sec. We have checked the sign of the rotation to be in agreement with the sign of the g^* factor.

The discrepancy between experimental results and theory (1) for high fields is not surprising because the Zeeman splitting between the Γ_{5+} and Γ_{5-} level is about 0.2 meV for 50 kG. This is comparable to the exchange-energy splitting (0.12

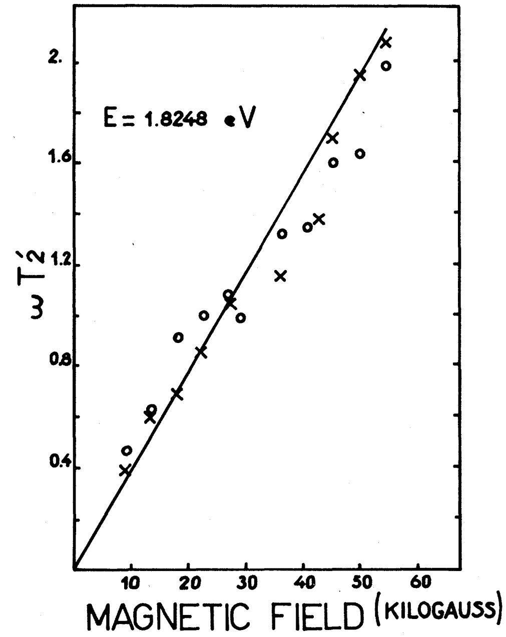

FIG. 2. From Eq. (1) in the text, one sees that the quantity $\omega T_2'$ is obtained, for a given magnetic field, (a) from the polarization rotation angle (crosses) $\omega T_2' = \tan 2\alpha$; (b) from the rate of polarization (circles) $\omega T_2' = [(P_0/P)^2 - 1]^{1/2}$. The plot shows a good agreement of experiment with theory for the 1.8248 resonant peak: The two determinations of $\omega T_2'$ are concordant and this quantity is linear in magnetic field. For other energies off the resonant peak, discrepancies occur for fields over 50 kG.

meV[11]). From another, but complementary, point of view, interferences may occur at the level crossing between the Γ_6 and Γ_5 Zeeman levels, and thus explain the spectral dependence of the discrepancy.

The interpretation of the variation of the experimental T_2' along the bottleneck requires a global analysis of all the data (intensity, polarization, and magnetic field effects) relative to the resonant peaks as a function of their energy. We just wish to point out here the relevant facts in this problem. We are dealing, in these Raman-like peaks, with a nonequilibrium population of polaritons, with well-defined energy (energy width ~0.2 meV). Thus, this technique appears to be a very good scanning probe to study the bottleneck region of the dispersion curve.

As we observe high degrees of polarization, we may conclude from the classical phenomenological theory of depolarization[5] that $\tau \ll T_2$, so that T_2' is of the same order of magnitude as τ. Thus, the depolarization lifetime may be considered, in a first approach, as a measure of the average time spent in the crystal by the oriented polaritons. It consists of (i) the time for emission of two LO phonons, scattering the polaritons down the band to an energy ϵ lying in the bottleneck region. This time is very short (of the order of 10^{-13} sec)[12] compared to (ii) the escape time of bottleneck polaritons, which depends on the depth in the crystal at which they were created, and on their group velocity $v_g(\epsilon)$. For example $T_2'(\epsilon)$ becomes very small when the observed polaritons can only be photonlike; that is, outside of the bottleneck region. This explains why no rotation is observed here. On the contrary, one would expect that $T_2'(\epsilon)$ grows as the inverse group velocity at the same energy:

$$T_2'(\epsilon) = d v_g(\epsilon)^{-1},$$

where d is the depth of absorption of incoming photons.

In fact, this growth is limited because of spin depolarization; in this model, the depolarization is due to acoustical phonon scattering of polaritons before their transmission at the crystal boundary.[3] Thus, a certain proportion of polaritons is observed after traveling in random directions of propagation, where the eigenstates (longitudinal and transverse) may not coincide with their state of polarization. This induces depolarization, which is a function of the energy-dependent acoustical scattering time $\tau_c(\epsilon)$. One deduces easily[10] that $T_2'(\epsilon)$ may be expressed, in a first approach, by

$$T_2'(\epsilon)^{-1} = d^{-1} v_g(\epsilon) + \tau_c(\epsilon)^{-1}. \qquad (2)$$

The maximum value we have found for T_2' is a good order of magnitude for the two terms involved in (2), when ϵ lies in the bottleneck region. This suggests that both must be considered in order to account for the spectral dependence of T_2'.

The results reported here show the importance of the polariton point of view in this problem: On one hand, the observed excitations of the crystal can be polarized and are subject to a kind of Faraday effect under longitudinal magnetic field. Their dependence upon exciting energy is very similar to resonant Raman scattering. On the other hand, the time spent in the crystal by these excitations can be much longer than is possible for ordinary photons. The associated energy indetermination (0.1 meV) is smaller than the longitudinal-transverse splitting (0.95 meV),[11] which shows that the exciton-photon interaction "has time" to be efficient.

Our method, in contrast to the ordinary Hanle effect, is based on an effect linear with the magnetic field and so allows the direct measurement of very short lifetimes. This is of interest in studying the scattering processes in the polariton bottleneck.

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Transferability of Phase Shifts in Extended X-Ray Absorption Fine Structure*

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Phase shifts in extended x-ray-absorption fine-structure (EXAFS) measurements have been empirically determined for atom pairs. For photoelectron energies >100 eV it is shown that these phase shifts, because they are essentially independent of chemical environment, can be used with EXAFS spectra to determine interatomic distances typically to accuracies of 0.02 Å.

Except for the case of an isolated atom, the x-ray absorption coefficient above threshold will be modulated as a result of interference between photoexcited outgoing electron waves and those same waves that have been backscattered from nearby neighboring atoms.[1] In a single-distance system this modulation, called the extended x-ray-absorption fine structure (EXAFS), is determined by two quantities—the interatomic distance and the phase shift describing the scattering effects from the absorbing and neighboring atoms. *The accuracy of interatomic distance determinations using the EXAFS technique is therefore inherently limited by the accuracy to which the phase shift is known. Ab initio* calculations of this quantity,[2-4] while encouraging, have been only qualitatively successful. In this Letter we report the empirical determination of phase shifts for atom pairs. The results are used to test the concept of chemical transferability: Phase shifts are sufficiently insensitive to chemical environment so as to be transferable from one system to the next.[5] The rationale for transferability is that for electron energies above 100 eV the scattering process is dominated by the core electrons, which are essentially unaffected by changes in the chemical environment. For the first time we shall show that transferability enables phase shifts from one system to be used for very accurate distance determinations in another system. Such a general procedure[5] is essential if the EXAFS technique is to make a significant contribution to structure studies; without it, accuracies better than 0.1 Å would be difficult, if not impossible, to achieve.

The EXAFS χ is defined as the normalized dif-