Observation of the Resonant Optical Stark Effect in a Semiconductor

D. Fröhlich, A. Nöthe, and K. Reimann
Institut für Physik, Universität Dortmund, 4600 Dortmund 50, Federal Republic of Germany
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This Letter presents for the first time the observation of the resonant optical Stark effect in a semiconductor. The excitonic system of Cu_2O is well suited to the investigation of this effect because of its rather large binding energy and well-resolved resonances at low temperatures. The mixing between the $1S$ and $2P$ excitons by the electric field of a tunable high-power CO_2 laser leads to drastic changes in the one-photon spectrum around the $2P$ absorption line. Even details of the line shape are consistently described in terms of the frequency-dependent nonlinear susceptibility.

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The investigation of the optical Stark effect has gained a lot of interest since high-power tunable lasers became available. Very recently Huo, Gross, and McKenzie[1] have reported the observation of the optical Stark effect in a molecular system. The observation of such an effect in solids, however, seems to be very difficult, because the linewidths of electronic transitions in solids are in general much larger than in atoms and molecules. This reduces the achievable resonance enhancement. On the other hand, the exciton system of a semiconductor offers good chances to observe the dynamical Stark effect, since large electric dipole matrix elements between appropriate states (e.g., $1S$ and $2P$) are expected. In this Letter we report the first observation of the resonant optical Stark effect in a semiconductor. The $1S$ and $2P$ exciton states of the well-known yellow exciton series[2] of Cu_2O are ideally suited to the demonstration of this effect because narrow resonances lines of these states are resolved with a tunable dye laser. More important, the energy difference between the $2P$ and $1S$ exciton states can be covered by a tunable CO_2 laser. The drastic changes in the dye spectra around the $2P$ resonance which are induced by the dynamical coupling of the $2P$ and $1S$ exciton by the high-power CO_2 laser can be explained quantitatively. Even details of the line shape are then consistently described in terms of the frequency-dependent nonlinear susceptibility χ^3.

Our measurements were performed on a highly pure arc-image-grown Cu_2O crystal. The sample of about 60-μm thickness was immersed in superfluid helium at 1.8 K. In our optical setup, which is described in detail by Fröhlich, Nöthe, and Reimann,[3] we used a transversely excited atmosphere CO_2 laser, which was tuned by a grating, and a tunable dye laser (rhodamine 6G pumped by an argon laser). The pulses of the CO_2 laser were shortened to about 20 ns by the use of a GaAs Pockels cell. This was necessary in order to suppress thermally induced effects due to a small absorption of the high-intensity CO_2 laser. At the sample the intensity of the CO_2 laser was about 100 $kW\,cm^{-2}$, which corresponds to an electric field of 5 $kV\,cm^{-1}$.

In Fig. 1 the coupling of the different laser fields to the exciton system is shown schematically. The low-power dye laser is tuned through the spectral region of the odd-parity $2P$ exciton. The $2P$ exciton is dynamically coupled to the even-parity $1S$ exciton by the high-power CO_2 laser (photon energy $\hbar\omega$), which can be tuned to change the resonance parameter $\Delta E = \hbar\omega - (E_{2P} - E_{1S})$. Without the CO_2 laser pulse we get one absorption band for $E = E_{2P}$. For the theoretical treatment of the exciton system, which interacts with the alternating field of frequency ω from the CO_2 laser, it is an excellent approximation to take into account only the $1S$ and $2P$ exciton states. The influence of all the other states (e.g., $2S$ and $3P$) can be neglected, because they are much more off resonance and their oscillator strengths are much smaller. The new eigenstates are gained from the diagonalization of a 2×2 matrix.[4] This gives a shift of the $2P$ ex-

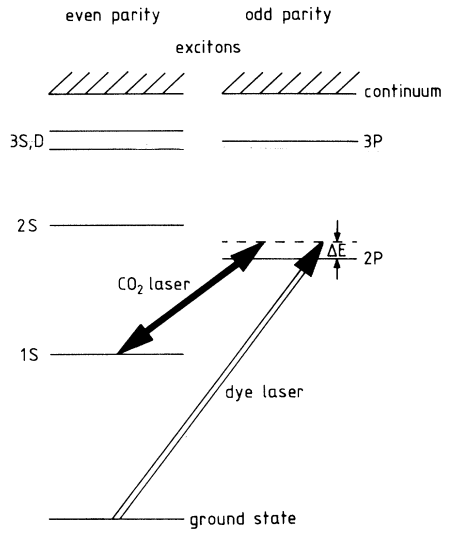

FIG. 1. Schematic level diagram of the yellow exciton series in Cu_2O. The $1S$ and $2P$ exciton states are coupled by the electric field of the CO_2 laser. The resulting states are probed by the dye laser. The detuning of the CO_2 laser from resonance is denoted by ΔE.

citon absorption to $E_2' = E_{2P} - \delta E$ and a new absorption at $E_1' = E_{1S} + \hbar\omega + \delta E$. The energy shift δE is given by

$$\delta E = \tfrac{1}{2}\{\mathrm{sgn}(\Delta E)[(\Delta E)^2 + 4V_{12}^2]^{1/2} - \Delta E\}. \quad (1)$$

In this equation V_{12} is the dipole matrix element between the $1S$ and $2P$ exciton states times the amplitude of the electric field. As stated above, the detuning parameter ΔE contains the frequency ω of the alternating field. The oscillator strength of the new absorption at E_1' is given by

$$f_1 = f_0 \sin^2[\tfrac{1}{2} \arctan (2V_{12}/\Delta E)], \quad (2)$$

where f_0 denotes the oscillator strength of the $2P$ exciton without an electric field. Accordingly the oscillator strength of the other state at E_2' is reduced to $f_2 = f_0 - f_1$.

The theory outlined above is only applicable in this form if the linewidths of the relevant states are much smaller than the detuning ΔE. In atoms and molecules this condition can be fulfilled close to resonance (small ΔE); in solids, however, only far off resonance. In the case of large linewidths and small detuning ΔE one has to integrate Eqs. (1) and (2) using the line-shape function of the excitons. From this analysis it follows that the states do not shift as a whole, but different parts shift by different amounts. The same applies to the oscillator strength. In order to resolve these effects we used a differential method, i.e., we measured the change in transmitted dye-laser intensity due to simultaneous interaction with the CO_2 laser radiation. The change of intensity was normalized to the intensities of the dye laser and the CO_2 laser. In Fig. 2 we present measurements for five values of the detuning parameter ΔE. A positive signal corresponds to an increase in absorption. For convenience the signal is plotted as a function of the difference between dye-laser and CO_2-laser energies ($E - \hbar\omega$). One can clearly see the drastic dependence of the signal on the detuning. The negative signal (decrease of absorption) is mainly due to a decrease in oscillator strength at this energy and in small part to the shift of the $2P$ exciton. At the high-energy side the positive signal is caused by the state at E_1', whereas the positive signal at the low-energy side stems from the shifted $2P$ exciton. In order to account for the detailed line shape of our experimental curves it is much more convenient to describe our results in terms of the nonlinear susceptibility[5] $\chi^{(3)}$ than to calculate the integrals over the line-shape function. With the normalization given above the signal is proportional to the imaginary parts of $\chi^{(3)}$. Because of resonance it is sufficient to retain only the dominant term,

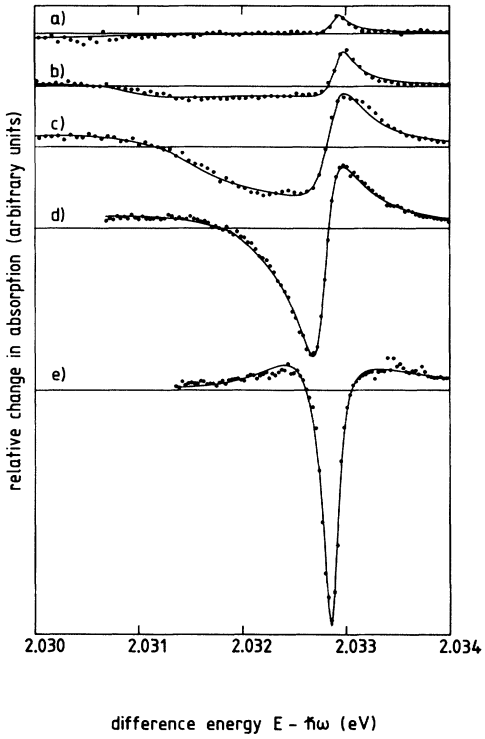

FIG. 2. Induced change in absorption as function of the difference of laser energies for five different energies of the CO_2 laser. The corresponding detuning parameters $\Delta E = \hbar\omega - (E_{2P} - E_{1S})$ are (a) 3.6 meV, (b) 2.3 meV, (c) 1.6 meV, (d) 0.9 meV, and (e) -0.08 meV. The dots represent the experimental results; the solid lines are the theoretical curves.

$$\chi^{(3)} \propto \frac{\langle 0|\hat{\mathbf{d}}\cdot\mathbf{r}|2P\rangle\langle 2P|\hat{\mathbf{c}}\cdot\mathbf{r}|1S\rangle\langle 1S|\hat{\mathbf{c}}\cdot\mathbf{r}|2P\rangle\langle 2P|\hat{\mathbf{d}}\cdot\mathbf{r}|0\rangle}{(E_{2P} - E - i\Gamma_{2P})^2[E_{1S} - (E - \hbar\omega) - i\Gamma_{1S}]}.$$

Here $\hat{\mathbf{c}}$ and $\hat{\mathbf{d}}$ are unit vectors in the polarization directions of the CO_2 and dye laser beams, respectively. The ground state is denoted by $|0\rangle$; $|1S\rangle$ and $|2P\rangle$ describe the $1S$ and $2P$ exciton states which have Γ_5^+ and Γ_4^- symmetry, respectively. For the theoretical fit in Fig. 2 the following parameters were used: $E_{1S} = 2.0329$ eV, $E_{2P} = 2.1479$ eV, $\Gamma_{1S} = 0.1$ meV, $\Gamma_{2P} = 1$ meV for $E > E_{2P}$, and $\Gamma_{2P} = 2.5$ meV for $E < E_{2P}$. The two values for Γ_{2P} take into account the pronounced asymmetry of the $2P$ exciton line. These parameters are taken from absorption data. Besides one scaling factor the theoretical fit requires no adjustable parameter. In Fig. 3 the experimental and theoretical values of the maxima and zeros of the signal are plotted as functions of the detuning parameter ΔE. No experimental values for the maxima on the low-energy side are included, since these maxima are

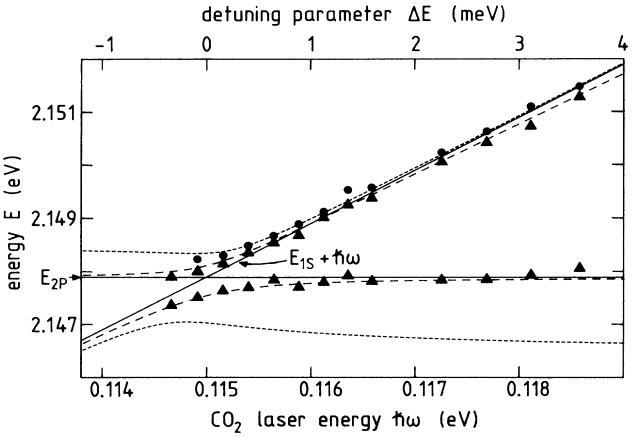

FIG. 3. Energies of the zeros and the maxima of the signal as functions of CO_2 laser energy and detuning parameter, respectively. The dots denote the experimental values of the zeros; the triangles are the values of the maxima. Short-dashed lines give the theoretical energies of the maxima, long-dashed lines the energies of the zeros.

rather broad, as can be seen in Fig. 2. Figures 2 and 3 show an excellent agreement between the experimental results and the theory.

One can think of other interesting experiments with this double resonant spectroscopy. Electronic systems should be investigated, where two sublevels—like the $1S$ and $2P$ exciton in Cu_2O—are strongly coupled by an electric dipole transition. Of special interest are multiple-quantum-well structures, where the electronic level structure can be tailored to fit the available laser energies. Time-resolved measurements should then be done, since they can yield information about relaxation processes in the different electronic levels.

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