Two-Photon Generation of Excitonic Molecules and Optical Bistability

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The fusion of two excited polaritons into an excitonic molecule gives rise to an intensity dependence of the dielectric function. The use of this nonlinearity to obtain optical bistability with platelets of direct–band-gap semiconductors is suggested.

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Optical bistability can be observed[1] in systems in which strong intensity-dependent changes of the real or imaginary part of the dielectric function occur. The considerable interest in this nonlinear optical phenomenon is due to its possible device application in integrated optics. Optical bistability can be described as a first-order nonequilibrium phase transition and the resulting photon distribution is expected to show anticorrelation effects. To generate optical bistability it was suggested recently[2] that one use the nonlinearity due to two-photon resonance lines. In highly excited semiconductors, a very efficient two-photon absorption process exists in which excitonic molecules are created via a nearly resonant excitonic level.[3]

It has been shown previously[4,5] that in the vicinity of the biexciton resonance the complex dielectric function is given by

$$\epsilon(\omega)=\epsilon_\infty+(\epsilon_0-\epsilon_\infty)\omega_x^2\left[(\omega_x^2-\omega^2)-\frac{4n_p|M|^2\omega_x(\omega_m-\omega-i\gamma_m)}{(\omega_m-i\gamma_m)(\omega_m-2\omega-i\gamma_m)}\right]^{-1}, \tag{1}$$

where ϵ_0 and ϵ_∞ are the static- and high-frequency dielectric constants; ω_x and ω_m are the frequencies of the exciton and biexciton, respectively; M is the matrix element for the biexciton generation; n_p is the polariton concentration; and γ_m is the effective width of the molecule state. The normal modes which result from (1) show an intensity-dependent dispersion around $\omega_m/2$. This dispersion has been observed by two-photon resonance Raman scattering in CuCl (Itoh and Suzuki[6]) and CdS (Kurtze *et al.*[7]). Equation (1) describes the observed intensity-dependent renormalizations quantitatively.[6] The corresponding reflection coefficient for normal incidence $R=[(n'-1)^2+n''^2]/[(n'+1)^2+n''^2]$ is plotted for two polariton concentrations in Fig. 1.

This intensity-dependent resonance in direct–band-gap semiconductors is well suited to generate optical bistability and may indeed be the mechanism for the recently observed optical bistability[8] in GaAs at a frequency just below the exciton peak.

The intensity which is transmitted through a Fabry-Perot resonator filled with an active medium is given by

$$\frac{J_t}{J_0}=\frac{(1-R)^2}{(e^{\delta''}-Re^{-\delta''})^2+4R\sin^2\delta'}, \tag{2}$$

where J_0 and J_t are the intensities of the normally incident and transmitted beams, respectively. $\delta=\delta'+i\delta''=Dn\omega/c$ is the phase shift for a single path through the resonator of width D. The frequency dependence of the complex phase shift is

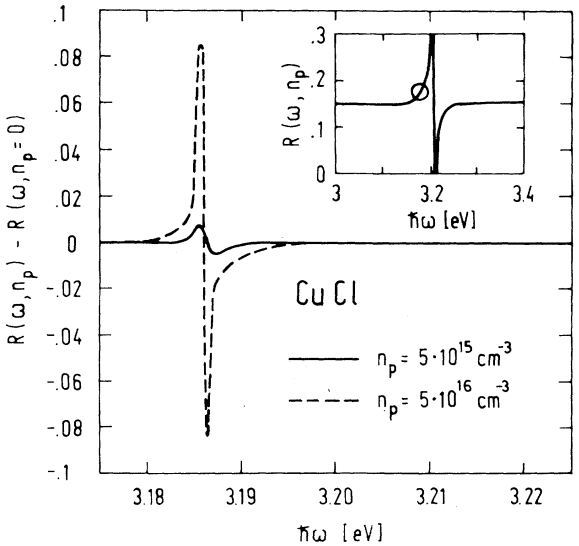

FIG. 1. Spectrum of the reflection coefficient $R(\omega, n_p)-R(\omega,0)$ of CuCl for two polariton concentrations. The inset shows the total exciton resonance and indicates by a circle at which frequencies the two-photon biexciton resonance occurs.

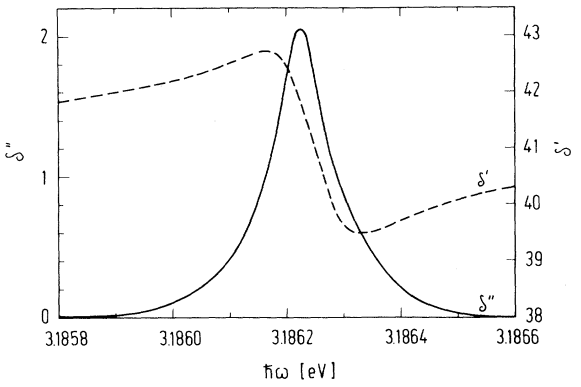

FIG. 2. Spectrum of the real and imaginary part of the phase shift (δ' and δ'', respectively) for CuCl with a thickness $D=1$ μm for a polariton concentration $n_p = 5\times10^{15}$ cm^{-3}.

plotted in Fig. 2. The intensity of the transmitted beam is related to the internal intensity J_i, which in turn can be expressed by the polariton concentration:

$$J_t \simeq (1-R)J_i/2, \qquad J_i \simeq n_p \hbar\omega v_g, \tag{3}$$

where $v_g(\omega)$ is the polariton group velocity.

Inserting Eqs. (3) into Eq. (2), one obtains a nonlinear equation for the polariton density n_p (or J_t). The resulting polariton densities are plotted in Fig. 3 as a function of J_0 and ω for the example of a platelet of CuCl which forms a resonator of 1 μm thickness. The used material parameters are those of Ref. 5, which have been shown to allow a quantitative fit of the intensity-dependent two-photon Raman spectra of Itoh and Suzuki.[6] The surface in Fig. 3 has a fold in which multiple solutions of n_p (and J_t) exist. Crossing the fold, e.g., by increasing the intensity J_0 of the incident beam at constant frequency, the polariton density will jump discontinuously from the lower to the upper branch. A comparison with Fig. 2 shows that the bistability which is shown in Fig. 3 is mainly a dispersive one.

The critical intensities of the incident beam $J_0 \simeq 0.1$ MW/cm^2 which we predict for the occurence of the optical bistability in CuCl platelets can be obtained easily with available uv dye lasers. Figure 3 shows that the bistability vanishes due to the increasing absorption when ω approaches the resonance at $\omega_m/2$. The bistability becomes more pronounced with decreasing ω but at the same time the necessary intensities become larger. We have chosen a rather small sample thickness D. Larger values of D will have the tendency to increase the necessary intensities due to increasing imaginary part δ'' of the phase shift. However, within one period of the resonator one can optimize the critical intensity by proper tuning. Another promising candidate for observing this optical bistability is CdS, for which the intensity-dependent dispersion due to the generation of excitonic molecules has also been observed recently in resonant two-photon Raman experiments[7] at excitation intensities which are even lower than those necessary for the corresponding experiment in CuCl.[6]

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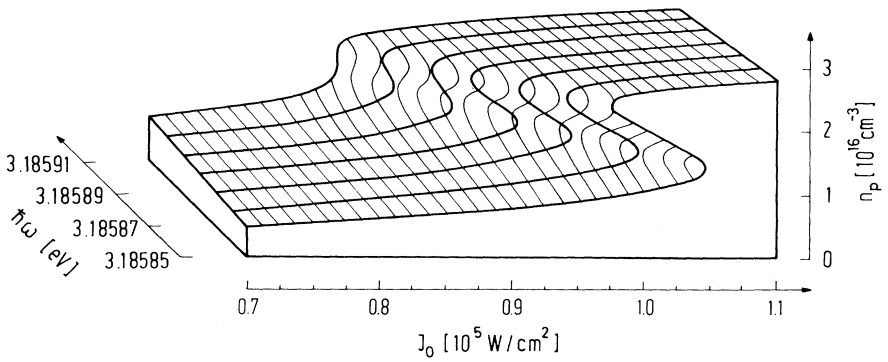

FIG. 3. The polariton concentration n_p in a CuCl resonator of 1 μm thickness as a function of the intensity and the frequency of the incident beam.

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Spin-Dependent Absorption of Electrons in a Ferromagnetic Metal

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It is found that the current collected by a ferromagnet placed in an electron beam depends on the orientation of the incident electron spin. At certain energies, only electrons with spins parallel or antiparallel to the net surface spin density cause a net target current. The spin dependence is caused by the influence of the exchange interaction on the elastic scattering. Inelastic scattering measurements show that the spin dependence of the production of secondary electrons is small.

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When low-energy electrons strike a metal, a variety of elastic and inelastic scattering phenomena occur. In the case of a ferromagnet, the interaction between the primary electron and the ordered net spin density of the sample electrons gives rise to a spin-dependent exchange interaction.[1,2] By using a primary beam of spin-polarized electrons and a ferromagnetic target, it is now possible to measure directly the effects of the exchange interaction in the elastic and inelastic channels. We present measurements to show that the effect of the exchange interaction is generally of the order of 10^{-2} of the spin-averaged interaction, for primary-electron energies E_0 of 2–500 eV. However, the exchange interaction can have a dominant effect on the net current absorbed by the sample at certain primary energies; either $i_a^{\uparrow\uparrow}$ or $i_a^{\uparrow\downarrow}$ can be finite while the other is zero, where i_a is the net absorbed electron current (number of electrons per second) and $\uparrow\uparrow$ ($\uparrow\downarrow$) means the polarization of the incident beam is parallel (antiparallel) to the majority-spin direction in the sample. To elucidate the mechanism behind this striking phenomenon, we present the first measurements of the spin-dependent asymmetry in inelastic scattering and secondary production. These suggest that the primary cause of the spin-dependent absorption is the spin-dependent interaction in elastic scattering. Through these results we demonstrate that polarized electron scattering presents a simple way to study various elastic and inelastic processes in a ferromagnetic electron gas and to obtain information on surface magnetic properties. Furthermore, the spin dependence of the absorbed current provides a new principle for detecting the spin polarization of an electron beam much superior to the complicated and inefficient methods in use or proposed.[3,4]

Spin-dependent electron scattering from a ferromagnetic surface was first measured by Celotta *et al.*[5] on Ni(110) with use of the spin-polarized electron beam emerging from a GaAs photocathode. In the present experiment, the spin-polarized electron beam is incident normal to the surface of the ferromagnetic glass, $Ni_{40}Fe_{40}B_{20}$. The electrons scattered from the sample are measured with a movable Faraday cup with an energy analyzing element to obtain the elastically scattered current $i_s(E_0)$ or the inelastically scattered current $i_s(E)$. The current absorbed by the sample, i_a, can be measured by a meter connected to the sample. An advantage of using a metallic glass is that it can be easily magnetized[6]; this leads to minimal stray magnetic fields outside the surface. The sample is a $16\times2\times0.03$ mm^3

