Nonlinear Spectroscopy of Biexcitons in CuCl by Resonant Coherent Scattering

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(Received 1 May 1978)

A new method of nonlinear spectroscopy aiming to probe solids in the highly dispersive and absorbing region is presented. It requires only one laser and uses wave-vector rather than frequency discrimination of the nonlinear signal from the input beams. Application is made to the study of the Γ_1 biexciton level in CuCl.

Recently multiphoton-active spectroscopy has been successfully applied to the study of atoms and condensed media,[1-3] providing information which cannot be obtained by linear spectroscopy. Most often the nonlinear optical process is a four-wave mixing whose efficiency is related to the dispersion of the relevant third-order susceptibility at certain combinations of the two laser frequencies $\omega_{1,2}$.[4-6] Generally these methods may be difficult to use in the highly dispersive region of a solid, i.e., when ω_1 or ω_2 lies close to an electronic resonance, because the phase-matching conditions may be impossible to fulfill or may give rise to complicated configurations because of polariton effects. In this Letter we wish to present a new method of active nonlinear spectroscopy aimed at probing solids well above their band gap. It provides information which is complementary to that given by two-photon absorption (TPA) and can be used in regions with very large absorption close to resonance, as shown below. It requires only one laser and uses wave-vector rather than frequency discrimination. Since all the optical beams have the same frequency, the phase-matching condition is rather insensitive to the dispersion of refractive indices. We have used these particularities to investigage the two-photon resonance of the biexciton level in CuCl. We first describe our method and present an analysis of the interaction in the parametric approximation, in order to reveal its salient features. Then some preliminary results relative to its application to CuCl are presented and discussed.

A laser beam is split into two parts, labeled "pump" and "test" beam, respectively. They are recombined at a very small angle θ_{ext} in a slab of material, with thickness l. The nonlinear interaction gives rise to a nonlinear polarization through a third-order process,

$$P_s(\omega)=\epsilon_0\chi^{(3)}(-\omega;\omega,\omega,-\omega)\vdots\vec{E}_p(\omega)\vec{E}_p(\omega)\vec{E}_t^*(\omega),$$

which in turn coherently radiates waves at frequency ω and wave vector $\vec{k}_s$. The wave-vector mismatch $\Delta\vec{k}=2\vec{k}_p-\vec{k}_t-\vec{k}_s$ reduces to its minimum value $|\Delta k|=n_0\omega\theta^2/c=(2\theta_{\text{ext}}^2/n_0\lambda)\pi$ for a direction of $\vec{k}_s$ symmetric to that of the test beam with respect to the pump one. The conversion efficiency, governed by $|\chi^{(3)}|^2\sin^2(\Delta k\,l/2)/(\Delta k\,l/2)^2$, is maximum in this direction so that a collimated beam is radiated by the sample at the angle $(-\theta_{\text{ext}})$. Its intensity is related to $\chi^{(3)}$ and becomes large whenever 2ω crosses a resonant energy level of the medium. An easy spatial discrimination is possible from $\theta_{\text{ext}}=1^\circ$ and with a thin sample the residual phase mismatch is unimportant [we have investigated the case $l=200$ μm, $\lambda\simeq0.4$ μm, $n_0\simeq2.5$ so that $\sin^2(\Delta k\,l/2)/(\Delta k\,l/2)^2\simeq0.95$].

In order to get a better description of this interaction we present now an analysis in the parametric approximation; i.e., the test beam is so weak that the pump depletion due to its interaction with the test and the signal beams is neglected but pump-induced two-photon absorption and optical Kerr effect are accounted for. We choose the z axis along $\vec{k}_p$ and only consider the variations along this direction. We assume the process to be exactly phase matched (a more detailed analysis will be presented elsewhere[7]); under those hypotheses the set of equations describing the interaction is

$$\partial E_p/\partial z+(\tfrac{1}{2}\alpha-\tfrac{1}{2}i\kappa I_p)E_p=0, \tag{1a}$$

$$\partial E_s^*/\partial z+(\tfrac{1}{2}\alpha+i\kappa^*I_p)E_s^*=-\tfrac{1}{2}i\kappa^*I_pE_t\exp[-2i\varphi(z)], \tag{1b}$$

$$\partial E_t/\partial z+(\tfrac{1}{2}\alpha-i\kappa I_p)E_t=\tfrac{1}{2}i\kappa I_pE_s^*\exp[2i\varphi(z)], \tag{1c}$$

where α is the absorption coefficient, $\kappa=\omega\chi^{(3)}/\epsilon_0n_0^2c^2$, $I_p=\frac{1}{2}\epsilon_0n_0c|E_p|^2$, and $\varphi(z)$ is the phase of the pump wave. The solution of Eq. (1a) is known; we have found an analytical solution for the whole system. Let us put $\kappa=\kappa_1+i\kappa_2$, $\Delta=|\kappa|^2-\kappa_1^2$, and $F=1+(\kappa_2 I_p^0/\alpha)[1-\exp(-\alpha z)]$; then

$$I_p(z)=I_p^0 e^{-\alpha z}/F, \quad \varphi(z)=(\kappa_1/2\kappa_2)\ln F, \tag{2a}$$

$$I_s(z)=(I_t^0 e^{-\alpha z}/F^2)(|\kappa|^2/\Delta)\sinh^2(\sqrt{\Delta}\ln(F)/2\kappa_2), \tag{2b}$$

$$I_t(z)=(I_t^0 e^{-\alpha z}/F^2)[1+(|\kappa|^2/\Delta)\sinh^2(\sqrt{\Delta}\ln(F)/2\kappa_2)]. \tag{2c}$$

Experimentally one can most easily measure three ratios: $\rho_1=I_s(l)/I_t(0)$, $\rho_2=I_s(l)/I_t(l)$, and $\rho_3=I_t(l)/I_t(0)$. Let us first remark that ρ_3 is the quantity customarily measured in two-beam TPA experiments; it is seen from (2c) that the usual analysis of TPA, which neglects the parametric gain experienced by the test beam in the case of substantial phase matching, is therefore doubtful. The ratio ρ_1 is related to both the modulus and the imaginary parts of $\chi^{(3)}$. It is important to notice that the factor $F^{-2}\exp(-\alpha l)$ which accounts for one- and two-photon absorption does not appear in ρ_2 because the two beams experience the same attenuation in the sample. Therefore ρ_2 is the relevant quantity to measure since it provides a means to observe resonances of $\chi^{(3)}$ around 2ω even if ω lies in a region of large absorption. This feature is most interesting to use for probing a solid close to the band gap. Indeed ρ_2 is mostly related to the modulus of $\chi^{(3)}$ as can be seen from its limit for small conversion or negligible TPA: $\rho_2\simeq[|\kappa|I_p^0\alpha^{-1}(1-e^{-\alpha l})]^2$. The comparison of ρ_1 and ρ_2 allows an absolute calibration of $\mathrm{Im}(\chi^{(3)})$ with respect to $|\chi^{(3)}|$.

An up-to-date review of biexciton-related topics can be found in the recent paper of Hanamura and Haug.[8] Extensive studies in CuCl by two-photon-induced luminescence[8,9] and absorption[10,11] have led to assignment to the Γ_1 biexciton of a narrow level at about 36 meV below twice the energy of the $Z_T{}^3$ exciton. In these experiments excitonic molecules are actually created in the crystal, and the intricate relaxation processes back to the ground state complicate somewhat the interpretation of luminescence experiments. In this respect coherent optical effects have the advantages of fixing both the initial and final states and yet providing information about energy states. To the best of our knowledge the only observation in CuCl of a coherent optical effect resonantly enhanced at about twice the exciton energy is the collinear four-wave mixing experiment recently reported by Mita and Nagasawa.[12]

In our experiment we use a N_2-laser–pumped dye laser, built according to the model described by Shoshan, Danon, and Oppenheim,[13] and followed by one amplifier. With the Lambda Physik's BbQ product, the dye laser can be tuned over the 3680–3920-Å range, with a 0.3-Å bandwidth and a peak power up to 6 kW. The laser beam is split into two parts, in the ratio 1/3, which are focused at a small angle on a 200-μm-thick plane-parallel slab of CuCl at the end of the cold finger of a liquid-He cryostat. The two laser beams are linearly polarized along a (001) axis of the crystal. The laser spot diameters on the sample are 200 μm.

Under these conditions a very strong resonance is observed when the laser frequency is in the vicinity of the so-called "biexciton giant two-photon resonance."[8] We have recorded the ratios ρ_1 and ρ_2 as a function of the frequency at moderate intensity $I_p^0\simeq 2.5$ MW/cm^2. For this intensity, and on the assumption that all the laser power is absorbed, the heating of the crystal is negligible (< 10 K). Typical recordings of ρ_1 and ρ_2 are shown in Figs. 1(a) and 1(b), respectively. The upper curves correspond to the two beams crossing in the sample. The lower ones are obtained by a slight misalignment (about 500 μm) of the beams, so as to let through the detection chain the background light existing when nonlinear optical mixing occurs. The resonance peaks are very unsymmetric. The minimum observed in ρ_1 occurs at 3891.5 Å and corresponds to the maximum two-photon absorption, in good agreement with the results of Grun and his co-workers.[11] The maximum of ρ_2 is observed at 3892.8 Å. It is shifted towards high energies with respect to that of ρ_1 by an amount of 1.5 Å.

The secondary hump at 3899 Å is indeed reproducible and corresponds to a dip in $I_+(l)$ at the same wavelength. The maximum of ρ_2 is 0.14 ± 0.02. The width of the resonance is significantly larger than expected both from TPA data[11] and the simple analysis above. This discrepancy is most likely attenuated if one includes in the analysis signal- and test-beam TPA and pump depletion. This requires numerical calculations which will be presented elsewhere.[7] Nevertheless, in order to be in the condition where the parametric approximation holds, we have recorded ρ_1 with attenuated test beams up to $I_t(0)/I_p(0)=1/30$. We did not observe essential changes in the overall shape of the spectra which remained unsymmet-

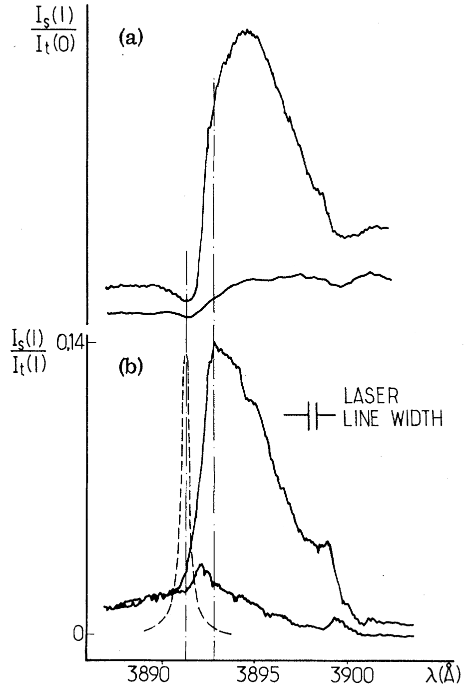

FIG. 1. (a) Upper curve, recording of the ratio $I_s(1)/I_t(0)$; lower curve, background (no overlap of the beams in the sample). (b) Upper curve, recording of the ratio $I_s(1)/I_t(1)$; lower curve, background (no overlap of the beams in the sample); dashed curve, shape of the resonance predicted by an isolated biexciton level, with normalized magnitude.

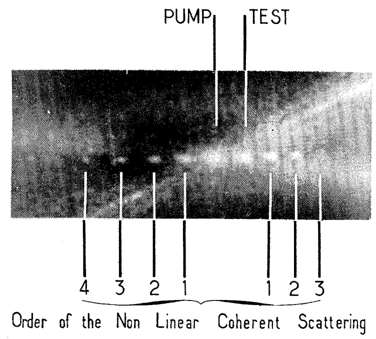

FIG. 2. Photograph of the beams emitted at resonance in a 200-μm-thick CuCl sample. The outer spots correspond to nonlinear coherent scattering up to fourth order; the two inner spots, to the pump and the test beams.

ric and broad; it is then necessary to consider the intensity dependence of line shapes, the correct description of damping,[14] and the autoionizing character of the biexciton state.[15]

The interaction is very nonlinear and at higher intensity ($I_P^0 \simeq 20$ MW/cm^2) the conversion efficiency is so large close to the resonance that up to four spots on the pump-beam side and three on the test-beam side could actually be seen to be emitted by the sample. This is shown in Fig. 2, which was obtained by photographing a white cardboard sheet intercepting the beams at about 15 cm from the sample. The magnitude of the effects is unexpectedly large for such a small interaction length! The spots correspond to the directions $\vec{k}_s^{\,n} = \vec{k}_p + n(\vec{k}_p - \vec{k}_t)$ and $\vec{k}_s^{\,n'} = \vec{k}_t + n'(\vec{k}_t - \vec{k}_p)$, respectively. The frequency dependence of the intensity of the high-order scattering beams is similar to that of the signal (first order) in the low-intensity experiment. At the maximum conversion the ratio of the intensities of the successive beams is fairly constant: $I_s^n/I_s^{n-1} \simeq 0.2 \pm 0.05$. The interaction is dominated by the two-photon resonance on the biexciton ($2\omega \sim \Omega_{bg}$) and the one-photon quasiresonance on the exciton ($\omega \sim \Omega_{eg}$); thus the $\chi^{(2n+1)}$ susceptibility may be written

$$\chi^{(2n+1)} = N\frac{(n+1)}{2^n}\frac{|\mu_{eg}|^2|\mu_{be}|^{2n}}{\hbar^{2n+1}(\Omega_{eg}-\omega)^{n+1}(\Omega_{bg}-2\omega+i\Gamma_b)^n}.$$

For the beam scattered in the k_s^n direction, the ratio of the nonlinearity driving the direct process to that involved in cascades of successive lowest-order scatterings depends only on the exciton linear susceptibility,

$$\chi^{(2n+1)}/(\chi^{(3)})^n = 2^{-n}(n+1)(\chi_{\rm ex}^{(1)})^{1-n},$$

where

$$\chi_{\rm ex}^{(1)} = N|\mu_{eg}|^2(\hbar\Omega_{eg} - \hbar\omega)^{-1} \simeq 1.51.$$

Let d be the effective interaction length, then the ratio of the intensity scattered directly to that originated by cascades is $[2^{-1}(n+1)(2\omega l\chi_{\rm ex}^{(1)}/n_0c)^{1-n}]^2$. The direct processes are negligible as soon as $d > \lambda$, which is the case at maximum conversion, i.e., 2.8 Å from the TPA resonance. Under these circumstances the ratio of successive scattered-beam intensities,

$$I_s^{n+1}/I_s^n = (4\pi l\chi^{(3)}/\epsilon_0 n_0^2 c\lambda)^2 I_p I_t,$$

is indeed constant. As an upper estimate of d one can take the reciprocal of the absorption coefficient at this wavelength: $\alpha \simeq 100$ cm^{-1}. Using this value, and correcting the experimental data for reflections on the sample as well as for spatial and temporal intensity distribution of the beams lead to $\chi^{(3)}(3894.3\ \text{Å}) \geq 3\times 10^{-9}$ esu, in fairly good agreement with the value 4.8×10^{-9}

esu deduced from the TPA data of Ref. 12.

In conclusion, we have presented a new method of nonlinear spectroscopy most suitable for the study of resonances in highly absorbing solids. It has been successfully applied to perform the first active nonlinear scattering on the Γ_1 biexciton of CuCl up to the fourth order. Our preliminary results show that the shape and the position of the resonance as observed by active nonlinear processes are significantly different from those revealed by TPA experiments.

We wish to thank Dr. J. B. Grun, Dr. R. Levy, and Dr. B. Hönerlage from the Laboratoire de Spectroscopie et d'Optique du Corps Solide of the University of Strasbourg for very fruitful discussions, and the service of chemistry of this laboratory for providing us with high-quality samples.

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Collisionless "Current-Channel" Tearing Modes

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Analytical and numerical studies of collisionless tearing modes that are wider than the "current channel" are presented. The $m=1$ type of inertial mode is shown to be strongly unstable for typical tokamak shear and β_e. Large spatial extension and large growth rate make it a possible candidate for explaining plasma disruption.

We report, in this Letter, analytical and numerical studies of tearing modes in a collisionless plasma. The modes we discuss are characterized by a mode width λ_w greater than the width of the electron layer or the "current channel" $x_e = |\omega/k_\parallel' v_e|$. Here $k_\parallel' = k_y/L_y$, k_y is the azimuthal mode number, L_s is the shear length, v_e is the electron thermal speed, and λ_w measures the region in which the parallel electric field is nonzero. Laval, Pellat, and Vuillemin[1] were the first to present an $m \geq 2$ type of current-channel tearing mode; their result is instructively discussed by Drake and Lee.[2] Later, Chen, Rutherford, and Tang[3] found trapped-particle modification to the mode. Although, these results are easily recovered in the appropriate limit of our dispersion relation, we emphasize here the $m=1$ type of inertial tearing mode first pointed out by Hazeltine and Strauss.[4] Our analytical dispersion relation, which has been verified in detail numerically, modifies the previous results for $\beta_e > m_e/m_i$. We also clarify its relationship to other instabilities. Most importantly, we confirm the potentially rapid growth and wide parameter range for instability of inertial tearing modes with $m=1$ character (large Δ'). Whenever it is consistent to treat the plasma in a collisionless approximation, this mode would be a serious candidate to explain plasma disruption because of its large spatial extension and large growth rate.

The slab geometry formulation of the electro-

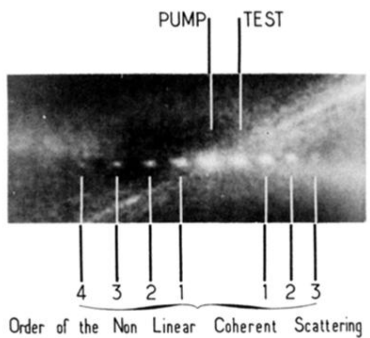

FIG. 2. Photograph of the beams emitted at resonance in a 200-μm-thick CuCl sample. The outer spots correspond to nonlinear coherent scattering up to fourth order; the two inner spots, to the pump and the test beams.