Interaction of light with toroidal oscillations in polar crystals

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The interaction of light with the density waves of the toroidal moment of band electrons is studied. The unusual spectral characteristics of the absorption coefficient of light in $TlGaSe_2$ is explained. An anomalous change of these spectral characteristics in the magnetic field is predicted. A possible detection of these features in antiferromagnets with local moments is discussed.

1. In the present letter we examine the optical properties of crystals which are similar to those of a toroidal transition.[2] A toroidal phase transition is characterized by the appearance of a peculiar antiferromagnetic order of the band electrons with a nonvanishing toroidal moment **T**[3]. In classical electrodynamics the toroidal moments are introduced as an independent family of multipoles in the expansion of the transverse density of the current $\mathbf{j}(\mathbf{r},t)$.[3] The toroidal dipole moment **T**, called below simply the toroidal moment, is given by

$$\mathbf{T} = \frac{1}{10c} \int \{\mathbf{r}\,(\mathbf{j}\mathbf{r}) - 2r^2\mathbf{j}\}\, d\mathbf{r} \ ,$$

where c is the speed of light. A transition to the toroidal state may be related to a softening of a particular mode of the collective electronic oscillations in a crystal.[4] This mode may interact with light in the infrared spectrum. Consequently, as the toroidal-instability point is approached, features may be seen in the spectral characteristics of the system and, in particular, in the optical-absorption coefficient $K_{\mathrm{T}}(\omega)$. We will show that because of the specific nature of the interaction of the toroidal moment with the electromagnetic field,[3] the behavior of $K_{\mathrm{T}}(\omega)$ near the toroidal transition point differs radically from an analogous functional dependence $K_{\mathrm{ph}}(\omega)$ in the scattering of light by a soft phonon mode in the case of a structural phase transition. Furthermore, the functional dependence $K_{\mathrm{T}}(\omega)$ changes markedly in a magnetic field, allowing the toroidal oscillations to be singled out in the case of complex spectral characteristics.

2. The Lagrange function of a system in which the low-frequency toroidal oscillations interact with the electromagnetic field is given by

$$\mathcal{L}_{\mathrm{T}} = K - U \tag{1}$$

$$K = \frac{(\dot{\mathbf{T}})^2}{2M_{\mathrm{T}}}\ , \qquad U = \alpha_{\mathrm{T}}\mathbf{T}^2 + \xi\mathbf{T}\dot{\mathbf{E}}\ , \tag{2}$$

where α_{T}, M_{T}, and $\xi > 0$ are coefficients which are analyzed in the microscopic model (see Ref. 2, for example), **T** is the density of the toroidal moment, and **E** is the electric field intensity. The unusual spectral dependence of the dielectric susceptibility, $\chi(\omega)$, is attributable in the experiments of Volkov *et al.*[2] to the particular way in which the toroidal moment **T** interacts with the field **E**. Varying (1) with respect to **T**, we find a dynamic equation of motion,

$$-\frac{\ddot{\mathbf{T}}}{2M_{\mathbf{T}}}-\alpha_{\mathbf{T}}\mathbf{T}-\gamma_{\mathbf{T}}\dot{\mathbf{T}}=\frac{\xi}{2}\dot{\mathbf{E}}. \tag{3}$$

Here we have added on the left side of (3) a term $\gamma_{\mathbf{T}}\dot{\mathbf{T}}$ which describes the damping in a system of toroidal moments (a microscopic justification is given in Ref. 4, in which $\vec{\pi}$ is used instead of **T**). Using (2) and (3), we find

$$\chi(\omega)=-\frac{1}{2}\,\frac{\xi^2\omega^2}{\dfrac{\omega^2}{2M_{\mathbf{T}}}-\alpha_{\mathbf{T}}-i\gamma_{\mathbf{T}}\omega}\,. \tag{4}$$

Here we have used the expression for polarization of the system $\mathbf{P}=\xi\dot{\mathbf{T}}$ derived from (2). The electrical properties of the nonstationary toroidal moment were discussed in detail in Ref. 5.

The absorption coefficient $K_{\mathbf{T}}(\omega)$ is proportional to $\operatorname{Im}\chi(\omega)$, and hence can be written as

$$K_{\mathbf{T}}(\omega)\sim\frac{\omega^3}{(\omega^2-\Omega_{\mathbf{T}}^2)^2+\lambda_{\mathbf{T}}^2\omega^2}, \tag{5}$$

$$\lambda_{\mathbf{T}}=2M_{\mathbf{T}}\gamma_{\mathbf{T}},\qquad \Omega_{\mathbf{T}}^2=2M_{\mathbf{T}}\alpha_{\mathbf{T}}.$$

A unique feature of expressions (4) and (5) is the unusual dependence of the numerator on the frequency of light, ω. In the case of ordinary polar oscillations, the oscillator strengths do not depend on the frequency and $\chi(\omega)\sim(\omega^2-\Omega_{\mathrm{ph}}^2-i\gamma_{\mathrm{ph}}\omega)^{-1}$.

The maximum of $K_{\mathbf{T}}(\omega)$ can be determined from the condition $\partial K_{\mathbf{T}}/\partial\omega=0$ which gives

$$\omega_{\mathbf{T}\max}^2(\Omega_{\mathbf{T}})=\left[\left(\Omega_{\mathbf{T}}^2-\frac{\lambda_{\mathbf{T}}^2}{2}\right)^2+3\Omega_{\mathbf{T}}^4\right]^{1/2}-\left(\Omega_{\mathbf{T}}^2-\frac{\lambda_{\mathbf{T}}^2}{2}\right). \tag{6}$$

The functional dependence $\omega_{\mathbf{T}\max}^2(\Omega_{\mathbf{T}})$ is illustrated in Fig. 1. We see that $\omega_{\mathbf{T}\max}^2\neq0$ for any value of $\Omega_{\mathbf{T}}$, including $\Omega_{\mathbf{T}}\to0\,[\omega_{\mathbf{T}\max}^2(0)\to\lambda_{\mathbf{T}}^2]$. The smallest value of $\omega_{\mathbf{T}\max}^2$ is reached at $\Omega_{\mathbf{T}}^2=\lambda_{\mathbf{T}}^2/4$ and $\omega_{\mathbf{T}\max}^2(\lambda_{\mathbf{T}}/2)=(3/4)\lambda_{\mathbf{T}}^2$. If $\Omega_{\mathbf{T}}\gg\lambda_{\mathbf{T}}$, then $\omega_{\mathbf{T}\max}^2\simeq\Omega_{\mathbf{T}}^2$ and is proportional to the soft-mode frequency. Accordingly, as $\Omega_{\mathbf{T}}$ is reduced, the absorption peak of $K_{\mathbf{T}}(\omega)$ first is displaced toward the low-frequency region and then at $\Omega_{\mathbf{T}}<\lambda_{\mathbf{T}}/2$ begins to move in the opposite direction. Such a behavior of $K(\omega)$ was observed in the $TlGaSe_2$ crystal[1] for one of the soft oscillation modes. The situation was complicated by the presence of another soft mode, for which the frequency $\omega_{\mathrm{ph}\max}$

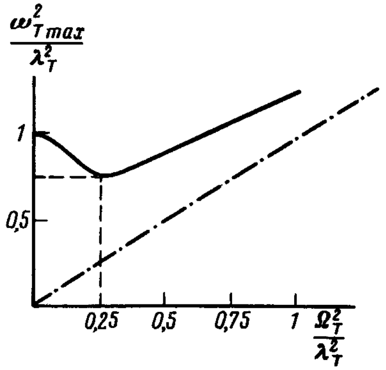

FIG. 1.

decreased monotonically with decreasing temperature. It can be assumed that $TlGaSe_2$ undergoes a structural phase transition at which the phonon mode softens ($\omega_{\mathrm{ph\,max}} \to 0$ as $\mathbf{T} \to \mathbf{T}_c$ and the frequency of the toroidal mode remains low but finite, $\Omega_\mathrm{T} \sim \lambda_\mathrm{T}/2$). It would thus be useful to trace the behavior of this mode below the point of the structural transition at $\mathbf{T} \ll \mathbf{T}_c$. Note that near the transition point the phonon and toroidal modes intermingle in accordance with the invariant $\mathbf{T}\dot{\mathbf{P}}_\mathrm{ph}$, where $\mathbf{P}_\mathrm{ph}$ is the polarization associated with the lattice vibration. We can therefore expect that these modes have an appreciable effect on each other if the frequency of the soft phonon mode, Ω_ph, is very close to the frequency Ω_T. The applicability of Lagrangian (1) and expression (4) is also limited by the low-frequency region, $\omega \ll E_g$, where E_g is the scale energy of single-electron excitations, which is on the order of the width of the energy gap of a semiconductor.

3. The functional dependence $K_\mathrm{T}(\omega)$ may vary appreciably in a magnetic field **H**. Adding the term[4]

$$\Delta U = \eta T(\mathbf{E} \times \mathbf{H}) \tag{7}$$

to (1) and performing several calculations, we find

$$\chi_\perp(\omega) = -\frac{M_\mathrm{T}[\xi^2\omega^2 + \eta^2 H^2]}{\omega^2 - \Omega_\mathrm{T}^2 - i\gamma_\mathrm{T}\omega}, \quad \mathbf{E} \perp \mathbf{H}. \tag{8}$$

For $\mathbf{E} \| \mathbf{H}$ Eq. (5) remains the same. Avoiding cumbersome calculations of $K_\mathrm{T}(\omega)$ for $\mathbf{E} \perp \mathbf{H}$, we point out that as **H** is raised, the minimum of $\omega^2_{\mathrm{T\,max}}(\Omega_\mathrm{T})$ shifts toward small Ω_T and when $|\eta H_0| \sim |\xi\gamma_\mathrm{T}|$ it vanishes. In a semiconductor model such as that in Ref. 2 we have

$$\xi \sim \frac{eP_{12}}{mE_g^2}, \quad \eta \sim \frac{e^2P_{12}}{cm^*mE_g^2},$$
$$M_\mathrm{T} \sim E_g^2, \tag{9}$$
$$\mu_B H_0 \sim \frac{m^*}{m}\gamma_\mathrm{T};$$

i.e., for $m^*/m \lesssim 10^{-1}$ we have $\gamma_T \sim \Omega_T \sim 10^{-2}$ eV and $H_0 \lesssim 100$ kOe. Here $\mathbf{P}_{12}$ is an interband matrix element of the momentum, m^* is the effective mass, m is the electron mass, E_g is the width of the energy gap, and μ_B is the Bohr magneton. The toroidal oscillation mode may thus be possible to identify even at $H \ll H_0$ from the change in $K_T(\omega)$ in a magnetic field at various orientations of the light-polarization vector. It would be useful to carry out relevant measurements of $TlGaSe_2$.

A doping with dipole impurity may lead to a suppression of the ferroelectric transition and render the toroidal transition more desirable. The minimum of $K_T(\omega)$ will be even more pronounced in this case and the static dielectric susceptibility $\chi_\perp(0)$ will diverge in a magnetic field:

$$\chi_\perp(0) = \frac{M_T \eta^2 H^2}{\Omega_T^2} \to \infty, \quad \Omega_T \to 0. \tag{10}$$

At the same time, $\chi_\parallel(0) = 0$ and does not have any anomalies at the toroidal-transition point.

4. These optical characteristics may also be found in antiferromagnets with localized moments if the magnetic-symmetry group of the crystal is consistent with invariants of the type $T_i L_j (i, j = x,y,z$, and $\mathbf{L}$ is the antiferromagnetism vector). The toroidal moment of the band electrons $\mathbf{T}$, in this case is induced by the spin-orbit interaction and the invariant such as $\mathbf{L}\dot{\mathbf{E}}$ in the functional of the antiferromagnet contains a relativistic small term.

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