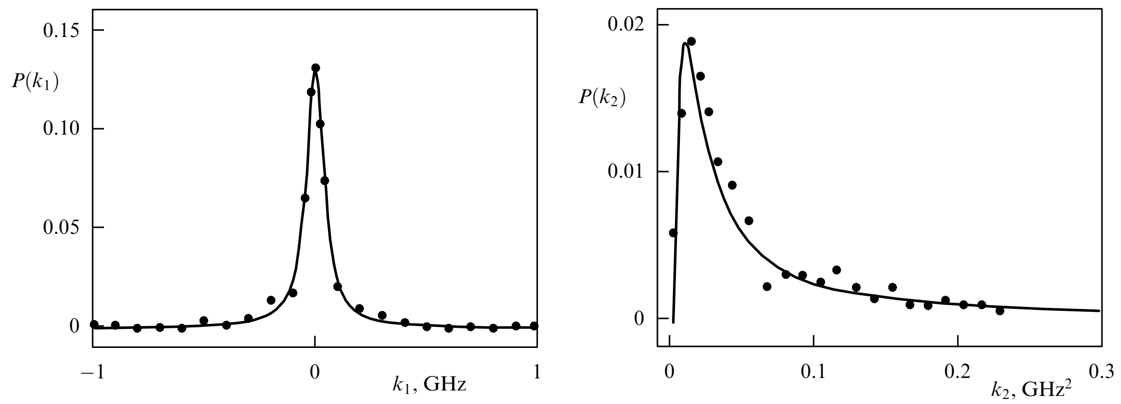

Figure 3. Distributions of the first and second cumulants obtained from the measured single-molecule spectra (dots) and their approximation by Lorentz and Smirnov functions (solid curves) predicted by the Levy statistics.

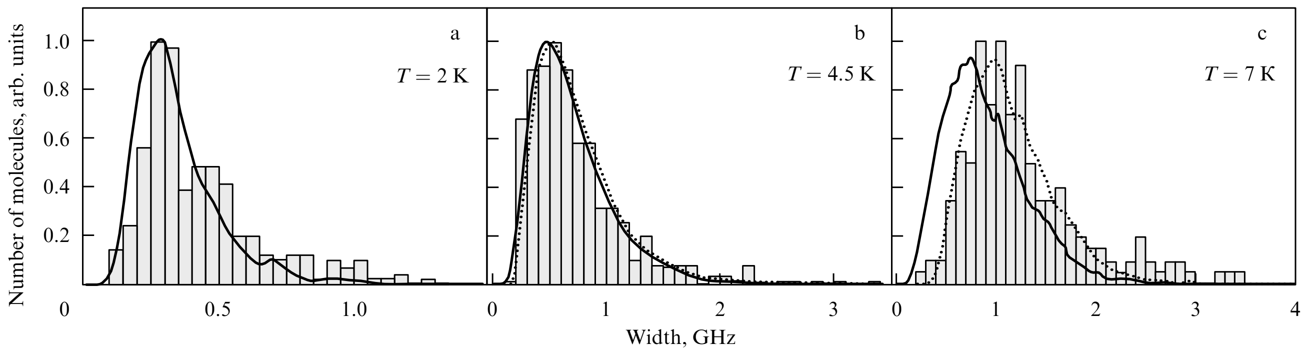

Figure 4. Experimental (histograms) and calculated (solid curves) peak width distributions in the spectra of single tetra-tert-butylterrylene molecules immersed into amorphous polyisobutylene. Dotted lines show model computation results in case of broadening caused by low-frequency modes of the matrix.

The distributions thus obtained are represented in Fig. 4 in the form of histograms. To compare the theoretical and measured SM spectra, we performed model calculations of SM spectra for the system under study and calculated their peak width distributions. The calculated distributions are depicted in the figure as solid and dotted curves. The solid curves were computed in the framework of the standard low-temperature glass model without accounting for the LFM contribution. The dotted curves represent the same distributions shifted toward larger width values to obtain better agreement with the experimental findings. It was assumed that the contribution of LFM to the line broadening is the same for all spectra and that it can be disregarded at $T = 2$ K. In this case, the magnitude of the shift determines the LFM contribution to the line broadening at a given temperature. Thus estimated, the LFM contributions are $\Gamma_{\text{LFM}} = 0.04$ GHz at $T = 4.5$ and $\Gamma_{\text{LFM}} = 0.24$ GHz at $T = 7$.

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Negative refraction in the optical domain and nonlinear wave propagation

V M Agranovich

Over 30 years ago, V Veselago noted very unusual properties of electromagnetic wave propagation in an isotropic medium with negative values of the dielectric permittivity and magnetic permeability, $\varepsilon < 0$, $\mu < 0$. Because the wave vector **K**, the electric field **E**, and the magnetic field **H** form a left-handed orthogonal set in such media, as opposed to the right-handed one formed in ordinary media, the former media are called left-handed materials (LHMs) in the English literature, to distinguish them from the normal right-handed materials (RHMs). An interesting and unusual property of wave propagation in these media is the antiparallel directions of the wave vector **K** and the Poynting vector **S**. Moreover, a beam refracted at the RHM/LHM interface turns out to be located at the same side from the normal to the surface as the incident beam (the so-called negative refraction). For this reason, LHMs are often called negative-refraction materials (NRMs).

Veselago's prediction has recently stimulated much theoretical interest and extensive experimental studies in search of new LHM (or NRM) materials [1–12]. Experimental success has been demonstrated in the microwave region [3, 6, 7]. Subsequently, however, it was shown that photonic-gap materials can also exhibit negative refraction. Similarly to the Bloch electron waves in crystals, optical waves in the periodic lattice of photonic-gap materials can have states with the opposite directions of the wave vector and the group velocity [8–12]. Negative refraction of light at the surface of a photonic crystal was demonstrated in a large number of numerical simulations [10–12].

The main attention in the studies on LHMs or NRMs has until recently been given to linear optical effects. In this report, we discuss nonlinear optical processes and show that they are also very unusual. We confine ourselves to the consideration of homogeneous NRMs and do not consider photonic-gap materials. The analysis of nonlinear optical processes in such materials is more difficult because it requires taking the optical Umklapp processes into account.

Before discussing nonlinear optical effects in NRMs, we note that two different approaches are typically used in studying the wave propagation.

One of them, based on the use of the Maxwell equations for the electric and magnetic fields **E** and **H** and the displacement vectors **B** and **D** (the so-called **E**-, **H**-, **B**-, **D**-picture) is usually applied to the investigation of the LHM electrodynamic properties. It is known, however, that this approach is applicable only in the low-frequency region because the magnetic dipole polarization density M loses its physical meaning in the high-frequency range [13]. The more general approach uses the **E**-, **B**-, **D**-picture in which $B = H$ with $\mu = 1$ and $D = \varepsilon E$, where the dielectric permittivity ε contains the entire linear response. In this approach, the dielectric tensor $\varepsilon(\omega, \mathbf{k})$ is characterized by both frequency and spatial dispersion. It is easy to see that the two approaches lead to identical results for the microwave range, where the introduction of magnetic permeability is justified. At the same time, the use of the **E**-, **B**-, **D**-picture [14] allows tracing the rise of LHMs when not only the magnetic dipole polarization but also the dielectric quadrupole one is taken into account. Moreover, it allows consistently passing to the optical wave range. The vectors **E**, **B**, and **K** then form a right-handed set in any medium, and the only nontrivial property of the so-called LHM postulated in the works of Veselago is the negative wave group velocity; therefore, negative refraction of a wave is a natural consequence of its negative group velocity [15, 16]. We have found no principal limitations on the occurrence of negative refraction in the optical wavelength range. Using the **E**-, **B**-, **D**-approach, we have discovered a number of unusual properties in negative-refraction media that emerge in studies of nonlinear optical processes, such as harmonic generation, stimulated combination (Raman) scattering, and propagation of short pulses.

We illustrate this by considering the generation of harmonics as an example. Because LHMs are normally realized in a narrow frequency range, an incident wave that occurs in this range has harmonics belonging to the frequency range where the medium exhibits positive refraction. This accounts for unusual relations between the propagation directions of the incident wave and the associated harmonics. It turns out that the harmonics generated by incident light in NRMs carry a major part of their intensity in the direction opposite to that of the incident beam propagation rather than in the direction of clear space as is typical of the generation of harmonics in ordinary media.

To conclude, we emphasize that negative refraction in the optical wavelength range can be realized in molecular crystals in the exciton resonance range (with a negative effective mass of the exciton), in gyrotropic materials, and for surface polaritons in the presence of transitional layers [17].

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Laser isotope separation by IR multiphoton dissociation of molecules

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1. Introduction

The optical separation of isotopes based on isotopic shift of electron transitions in atoms and simple molecules in the visible and ultraviolet ranges was experimentally demonstrated as early as the 1920s–1930s. Although it was clear that infrared (IR) spectra of molecules may undergo a large isotopic shift, the practical use of this phenomenon for isotope separation was hampered by an important limitation consisting in a small change in the chemical activity of a molecule upon absorption of one IR photon, and hence a small value of the potential isotopic selectivity of an elementary process. This obstacle was overcome with the