Lorentzian Line Crossings in Forward-Scattered Light*

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The zero-field line crossing of the mercury isotope ^{204}Hg was studied in forward-scattered light, with the scattering atoms both in a foreign gas and in vacuum. With sufficient foreign-gas pressure, a Lorentzian shape was obtained, in contrast to the Gaussian shape found for atoms in vacuum. An approximate theoretical treatment gives a field dependence of the forward-scattered intensity in satisfactory agreement with the measurements. Line crossings of Zeeman components of different isotopes, contained in separate cells, are also discussed.

Polarized resonance radiation scattered by atoms in vacuum, and in a uniform external magnetic field, is found to vary in intensity at field strengths where certain Zeeman levels of the atomic excited state cross. In laterally scattered light these intensity variations are called level crossings; they have a Lorentz shape, and a width determined by the Landé g factor and the mean life of the excited level. Related effects in light scattered forward by the same atoms are called "line crossings."[1] Line crossings previously observed have a Gaussian line shape and a width determined by the g factor and the Doppler breadth of the resonance radiation scattered by the atoms.[1,2] In both cases the atomic excited state is a coherent mixture of Zeeman levels. The observed differences arise from coherence properties of the scattered light. Laterally scattered light is incoherent, but forward-scattered light has an intensity proportional to the square of the atom density N, characteristic of coherence in the light emitted by different atoms.

Level and line crossings have been observed under conditions of broad-band excitation, e.g., by light with a spectral width exceeding the width or separation of the excited levels. This broad-band excitation is requisite for atomic coherence. We describe here coherence effects in forward-scattered light observed under narrow-band excitation conditions. A Lorentz shape is observed, which is adequately described by an appropriate modification of the theory used to discuss line crossings.[2]

Figure 1 shows the apparatus used to investigate forward-scattered light; it differs basically from that previously employed[1,2] only in the tunable light source, which is a mercury resonance lamp mounted in a magnetic field.[3] The circularly polarized σ_+ and σ_- components of the resonance radiation have different frequencies (or wavelengths) because of the Zeeman effect of the atoms in the lamp. When these components pass through the quarter-wave plate they become linearly polarized in orthogonal directions. One of these spectral components is selected by the linear polarizer. If this light is forward scattered by atoms in the cell, a magnetic-field-dependent rotation of the polarization direction may permit passage through the analyzing polarizer to the photomultiplier tube detector; all other light is sharply attenuated.

In quartz cells with $L = 2$ cm, the vapor pressure of Hg at room temperature or below is sufficiently low that the "weak-scattering approximation,"[2] corresponding to single scattering in laterally scattered light, is adequate for most measurements. The intensity of forward-scattered light is then described by the integral over angular optical frequency k:

$$I(H) \propto \int_0^\infty \rho(k) F^*(H,k) F(H,k)\, dk, \tag{1}$$

where $\rho(k)$ is the spectral density of light emitted by the lamp and $F(H,k)$ is written in terms of the plasma dispersion function[4] $Z(x+iy)$ as

$$F(H,k) = Z\left(\frac{k - k_0 - \gamma H + \frac{1}{2} i\Gamma}{\Delta}\right) - Z\left(\frac{k - k_0 + \gamma H + \frac{1}{2} i\Gamma}{\Delta}\right).$$

The gyromagnetic ratio of the $^3P_1$ level of Hg is written γ, $2\Delta\sqrt{\ln 2}$ is the full width at half-maximum of the light scattered by atoms in vacuum, $\Gamma/2\pi$ is the reciprocal mean life of the level, and k_0 is the central frequency of the absorption line of the atom.

For atoms in vacuum, the distribution of scattered light is narrow compared with the corresponding distribution of radiation from the lamp. The approximation $\rho(k) = \text{const}$ is appropriate, yielding a predicted intensity distribution of detected forward-scattered light with Gaussian line shape[2]:

$$I(H) \approx I_0 \sqrt{\pi}\,(1 - e^{-2\gamma^2 H^2/\Delta^2}), \tag{2}$$

with I_0 a constant. If the cell is filled additionally

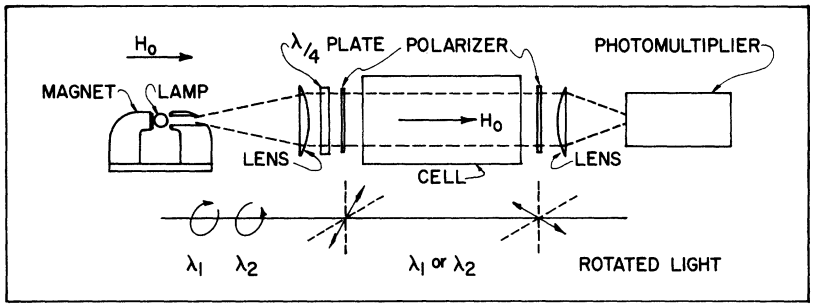

FIG. 1. Schematic diagram of the apparatus used in line-crossing measurements, and the polarization of the light in the optical system. The circularly polarized σ_+ and σ_- components were converted to orthogonal linearly polarized lines by the $\frac{1}{4}\lambda$ plate. Rotation of the plane of polarization in forward scattering permitted light to pass the analyzer. When the lamp was operated at zero field, no $\frac{1}{4}\lambda$ plate was used.

with an atmosphere of foreign gas, such as argon or N_2, there is a collision-induced broadening and shift of the mercury absorption.[5] At STP, Γ now describes the broadened width instead of the natural width, and $\Gamma >> \Delta$. The approximation $\rho(k) \approx \delta(k - k_0')$ is adequate for this case, k_0' being the center frequency of the lamp emission. Also, for $\Gamma >> \Delta$, $Z(x+iy) \approx -\Delta/(x+iy)$. If $k_0' = k_0$, the center frequency of the absorption, these approximations can be used in Eq. (1) to yield

$$I(H) \approx \frac{2C(kL/c)^2(2\pi^2 NP^2/h)^2\{1+[(\gamma H)^2 - \frac{1}{4}\Gamma^2/(\gamma H)^2 + \frac{1}{4}\Gamma^2]\}}{(\gamma H)^2 + \frac{1}{4}\Gamma^2}, \tag{3}$$

where P denotes the reduced matrix element of the intercombination transition of Hg at 2537 Å, C is a constant, and c and h are, respectively, the speed of light and Planck's constant. The shape is Lorentzian, but the final factor in parentheses in the numerator decreases the transmitted intensity when the Zeeman splitting exceeds the linewidth.

For this investigation, both a lamp and a cell containing ^{204}Hg were used, although small quantities of other isotopes were also present. Figure 2(a) shows observed transmitted intensity as a function of magnetic field strength applied to the cell, but with the lamp operated at zero field. The theoretical curves from Eqs. (2) and (3) are plotted in Fig. 2(b). The asymmetry of the zero-field line crossing for ^{204}Hg in vacuum [upper curve of Fig. 2(a)] is probably due to the presence of other isotopes. A decrease in transmitted intensity at high field strengths, not predicted by Eq. (2), occurs when the Zeeman splitting of the scattering atoms exceeds the spectral width of the lamp. One sees directly that the Lorentz broadening of the line crossing for Hg in 1 atm of argon [lower curve of Fig. 2(a)] is much greater than this spectral width. The Lorentz shift of the radiation scattered from isotope 204 in argon was partially compensated in the total broadened absorption curve by the presence of the small amounts of other isotopes. Consequently, the approximation $k_0' = k_0$ was adequate, if the absorption was treated as a single broadened line. No significant changes were observed when the lamp resonance was Zeeman shifted, as described above, to provide slightly better coincidence. Parameters used in the normalized fits to Eqs. (2) and (3) were $\Delta/2\pi = 1.3$ GHz, $\Gamma/2\pi = 21$ GHz, and $\gamma/2\pi = 2.1$ MHz/G. The values of Δ and Γ were obtained from the corresponding absorption curves. Neither fit in Fig. 2(b) is perfect in the region of validity, the small deviations from the data probably arising from the several approximations made.

Line crossings of Zeeman components of different isotopes[1] can also be studied at reduced foreign-gas pressures or in vacuum. Figure 3(a) shows the production of such a crossing of Zeeman components of isotopes 204 and 202 contained in separate evacuated cells. The ^{204}Hg lamp was used at zero field. Dashed curves show the normalized intensity of transmitted light for each cell alone; the solid curve shows the intensity transmitted with the two cells mounted in series in the light path, with the isotope 204 first in the light. The forward-scattered light was reduced in intensity by the additional quartz surfaces. The isotope shift separates the components at zero field by about 5.25 GHz,[6] so a crossing is expected in the solid curve near 1.25 kG, as well as at zero field. The consequence of reversing the order of the cells in the light path is shown in Fig. 3(b). Commutation is expected for optical rotations

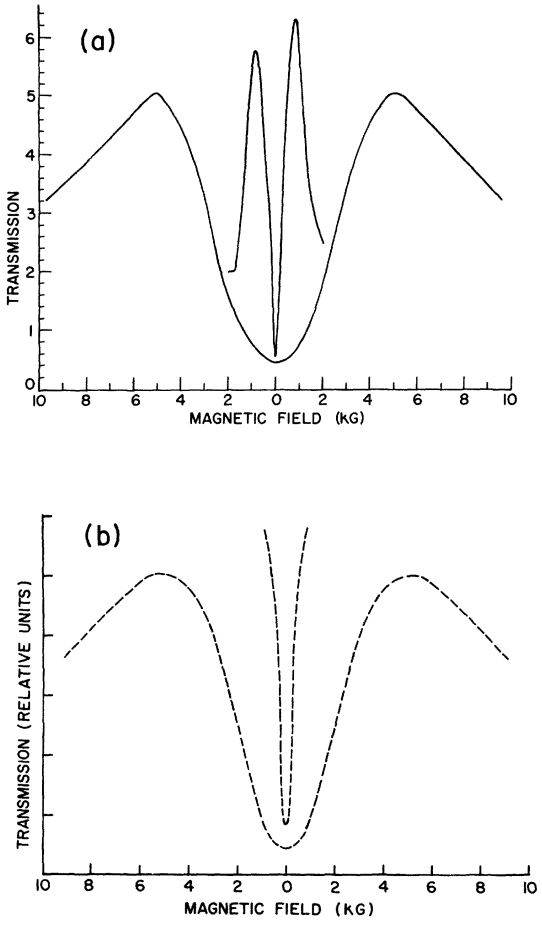

FIG. 2. (a) Zero-field line-crossing results for ^{204}Hg in vacuum (upper curve) and in 1 atm of argon (lower curve). (b) Curves plotted from Eq. (2) (upper curve) and Eq. (3) (lower curve), normalized to the observed light intensity, using parameters determined from absorption measurements (not shown).

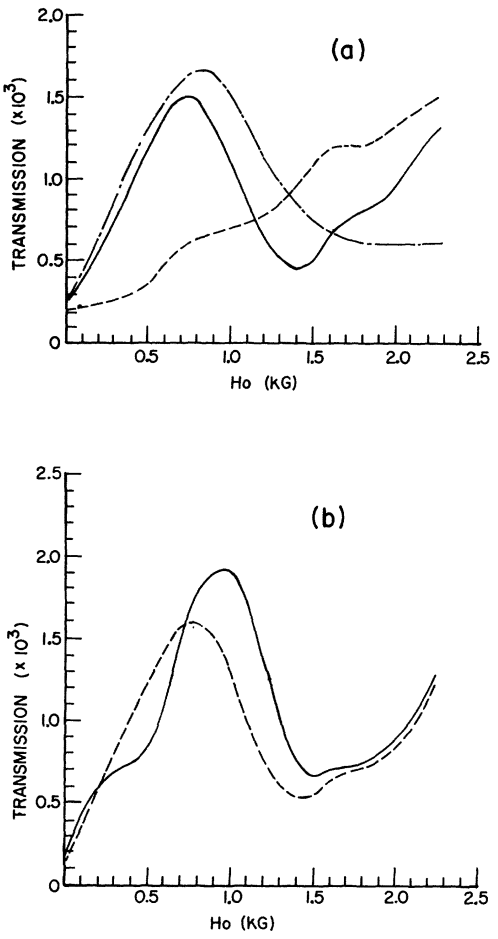

FIG. 3. (a) Forward-scattered intensity as a function of applied magnetic field strength for ^{204}Hg in vacuum (dash-dot line) and ^{202}Hg in vacuum (dashed line), and with the cells containing the two isotopes mounted in series in the light path (solid curve). (b) Forward-scattered intensity as a function of applied magnetic field strength for cells containing isotopes 204 and 202 mounted in series: isotope 204 first in the light beam (dashed curve); isotope 202 first in the light beam (solid curve). The isotopes were contained in separate evacuated cells.

about the same axis, so the effect is attributed to imperfect fulfillment of the approximations leading to Eq. (1), e.g., neglect of absorption. The appearance of the crossing minimum at slightly higher field than expected may also arise from this source. With both isotopes in the same cell, with one-half the total length of scattering vapor, the crossing minimum occurred at $H \approx 1.2$ kG. The isotopic densities were not in the same ratio as those in the previous measurements.

Lorentzian line crossings in forward-scattered light would appear to be useful for linewidth measurements in pressure-broadening studies. We join with others[1] to discourage the employment of line crossings for isotope-shift studies. Our primary motivation in this work was the desire to investigate coherent forward scattering as a means to detect the presence of trace quantities of pollutants in air. These further investigations will be reported elsewhere. The results to date indicate that basic research, in conjunction with applied research in atomic physics, can aid in the solution of current national problems.

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[1]R. Q. Hackett and G. W. Series, Opt. Commun. 2, 93 (1970).

[2]A. Corney, B. P. Kibble, and G. W. Series, Proc. R. Soc. Lond. A 293, 70 (1966).

[3]F. Bitter, Appl. Optics 1, 1 (1962); F. Bitter, H. Plotkin, B. Richter, A. Teriotdale, and J. F. R. Young, Phys. Rev. 91, 421 (1953).

[4]B. D. Fried and S. D. Conte, *The Plasma Dispersion Function* (Academic, New York, 1961).

[5]A. C. G. Mitchell and M. W. Zemansky, *Resonance Radiation and Excited Atoms* (Cambridge U. P., New York, 1961).

[6]W. G. Schweitzer, Jr., J. Opt. Soc. Am. 53, 1055 (1963).