Multiperturber effects in the Faraday spectrum of Rb atoms immersed in a high-density Xe gas

J. P. Woerdman, F. J. Blok,* M. Kristensen† and C. A. Schrama‡
Huygens Laboratory, Leiden University, P.O. Box 9504, 2300 RA Leiden, The Netherlands
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We have measured the D_1 and D_2 Faraday spectrum and absorption spectrum of Rb atoms immersed in high-density Xe buffer gas in the range $n_{Xe}=0.8-4.5\times10^{20}$ cm^{-3}. We find that the shape of the Faraday spectrum obeys the Becquerel relation over this whole density range; however the relative strength of the Faraday effect compared to absorption changes rather abruptly near $n_{Xe}=1\times10^{20}$ cm^{-3}. This is ascribed to the onset of a many-body nature (overlapping collisions) of the Rb:Xe line broadening; the number of perturbers within the Weisskopf sphere is unity at $n_{Xe}\sim1\times10^{20}$ cm^{-3}.

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I. INTRODUCTION

For a gas of free atoms the connection between spectral properties and collision physics is well established within the framework of spectral line-broadening theory. However, with increasing density this connection becomes highly nontrivial when quasimolecular conditions start to prevail, i.e., when the atoms are permanently in a state of collision. For instance, there is no practical way to calculate the optical properties of condensed matter on an *ab initio* basis; even brute force quantum chemical or band-structure calculations show in general poor performance. By studying the optical properties of a dense gas one explores, in a sense, the middle ground between atomic and condensed-matter physics. In this realm the use of line-broadening theory is questionable since this is generally based on the binary collision approximation. When the atoms are always in a state of collision, multiperturber interactions are dominant; generally, these interactions are not additive [1–3].

Our interest is, in particular, in *magneto*-optical spectroscopy as a probe of quasimolecular effects [4]. As in spectroscopy of free atoms, adding a magnetic field gives additional possibilities for studying structures. Recently, we have observed the onset of quasimolecular behavior in the core of the Faraday D_2 line of Rb atoms in Xe gas ($n_{Xe}\sim0.1-1\times10^{20}$ cm^{-3}) [5,6]. It was found that the results could be parametrized using standard, binary line-broadening theory despite the fact that the collisions were almost overlapping.

In the present paper we report an extension of the previous work to higher Xe densities ($n_{Xe}=0.8\times10^{20}-4.5\times10^{20}$ cm^{-3}). We observe a drastic change in the previously observed quasimolecular modification of the Faraday spectrum. This is apparently a consequence of the fact that we are now truly in the multiperturber regime and indicates that magneto-optical spectroscopy provides sensitive diagnostics of quasimolecular aspects. In the absence of a practical theory of nonbinary line broadening, the discussion of our experimental observations will be necessarily qualitative.

The structure of the paper is as follows. In Sec. II we discuss how violation of the so-called Becquerel relation acts as a signature of quasimolecular behavior. Section III is devoted to the experimental methods. In Sec. IV we report and discuss our experimental results. Section V gives concluding remarks.

II. VIOLATION OF THE BECQUEREL RELATION

Our diagnostics of the quasimolecular effects is based on a comparison of the Faraday spectrum and the dispersion spectrum. Briefly, when linearly polarized light passes through matter, the plane of polarization will rotate when a longitudinal magnetic field is applied (Faraday effect). The microscopic origin of the effect is that the magnetic-dipole moments of the atoms in the medium precess in the magnetic field. In a *dilute* atomic gas, the magnetic precession is unperturbed and the Faraday rotation angle θ_{Far} is proportional to the dispersion $dn/d\nu$, as expected from classical electrodynamics. This result is called the Becquerel relation [4–7],

$$\theta_{Becq}=\alpha_{Becq}\frac{e}{2mc}BL\nu\frac{dn}{d\nu}, \tag{1}$$

where e is the electron charge, m the electron mass, c the velocity of light, B the longitudinal magnetic field, L the length of the sample, and $\alpha_{Becq}=1$ for a spinless transition. In our experiment we deal with a gas of Rb atoms, which have both nuclear and electron spin. Equation (1) remains valid in the presence of spin if we deal with an isolated optical transition. In this case the lower and upper levels have well-defined g factors ($\neq1$), generally leading to $\alpha_{Becq}\neq1$ in Eq. (1). In the work reported here the linewidth is at least an order of magnitude larger than the Rb hyperfine splittings, so that the effects of the nuclear spin can be ignored. What remains is the effect of the Rb electron spin, i.e., the fine-structure splitting. This leads to $\alpha_{Becq}=\frac{4}{3}$ for the D_1 line and $\alpha_{Becq}=\frac{7}{6}$ for the D_2 line [4,7].

*Present address: Faculty of Technical Physics, University of Twente, P. O. Box 217, 7500 AE Enschede, The Netherlands.
‡Present address: NMI Laboratory, P. O. Box. 654, 2600 AR Delft, The Netherlands.
†Present address: Mikroelektronik Centret, DTU, Bldg 345 east,DK-2800 Lyngby, Denmark

We have predicted [4] that for an optical transition in a *dense* gas the Becquerel relation [Eq. (1)] is violated if the density is high enough, i.e., if the presence of nearby perturbers destroys the spherical symmetry of the potential felt by the Rb valence electron. To quantify what we mean by a "high" Xe density n_{Xe}, we introduce the Weisskopf number $N_W \equiv n_{\mathrm{Xe}}(\frac{4}{3}\pi R_W^3)$, with the Weisskopf radius $R_W=(\Delta\nu_{\mathrm{FWHM}}/n_{\mathrm{Xe}}v_r)^{1/2}$ as the impact parameter that yields unity optical phase shift in impact line broadening [$\Delta\nu_{\mathrm{FWHM}}$ is the full impact linewidth (full width at half maximum) and v_r the mean interatomic collision velocity]. The condition $N_W>1$ signals the importance of overlap of collisions. As an example, we calculate for the Rb D_2 line at 400 K that $N_W=0.84$ for $n_{\mathrm{Xe}}=1\times10^{20}$ cm^{-3}. Here we have used $\Delta\nu_{\mathrm{FWHM}}/n_{\mathrm{Xe}}=6.46\times10^{-10}$ s^{-1} cm^3 (this value can be deduced from the data reported in Ref. [6]) and $v_r=4.04\times10^4$ cm s^{-1} (at 400 K). In our previous work [5,6] we have observed violation of the Becquerel relation for the D_2 transition of Rb atoms immersed in Xe for $N_W\leqslant0.8$; in the present paper we report results for $0.8\leqslant N_W\leqslant4$.

III. EXPERIMENTAL TECHNIQUES

For a detailed description of the experimental setup, we refer to our previous work [6]; here we concentrate on some improvements. To gain access to the regime $N_W\geqslant1$ we have developed a Rb:Xe high-pressure cell. The final design was a 22-mm-long thick-walled glass tube (outside diameter 24 mm; inside diameter 4 mm) terminated by wedge-shaped (4°) windows, with a thickness of 10 mm, which were antireflection coated on both sides. The tube itself was also wedge shaped (1.3°). These precautions were taken to avoid étalon effects. The tube and the windows were made of Pyrex glass and were glued together with Silvac [8]. The cell was filled with Rb metal and Xe gas. The Rb vapor density (typically $N_{\mathrm{Rb}}\approx10^{12}$ cm^{-3}) was chosen by varying the cell temperature (typically 360–420 K) such that the absorption length at line center roughly equaled the cell length. Precise knowledge of the Rb density and its spatial distribution over the cell length is not required since it affects the Faraday rotation and optical absorption in the same way.

Using this cell we have measured the Faraday spectrum and the absorption spectrum of Rb:Xe for $n_{\mathrm{Xe}}=0.8$–4.5×10^{20} cm^{-3}. The dispersion spectrum $n(\nu)$ was derived from the absorption spectrum by Kramers-Kronig inversion. This allowed a quantitative check of the Becquerel relation [Eq. (1)]. The absorption spectra were recorded sufficiently far in the wings to enable accurate Kramers-Kronig inversion. The main difference as compared to our previous work is that we have used a tunable cw Ti:sapphire laser instead of a set of semiconductor lasers. This has allowed us to study not only the Rb D_2 line (780 nm), as before, but also the Rb D_1 line (795 nm). The linewidth of the Ti:sapphire laser was ~2 GHz, which is very small compared to the smallest Rb collisional linewidth encountered in the present work ($\Delta\nu_{\mathrm{FWHM}}\approx60$ GHz).

A key point is that an absolute comparison of the measured value of θ_{Far} and the indirectly obtained value of θ_{Becq} requires precise knowledge of the product BL [see Eq. (1)]. As before [5,6], we accurately determine BL by measuring the Faraday effect at "low" Xe density ($n_{\mathrm{Xe}}\sim3\times10^{18}$ cm^{-3}),

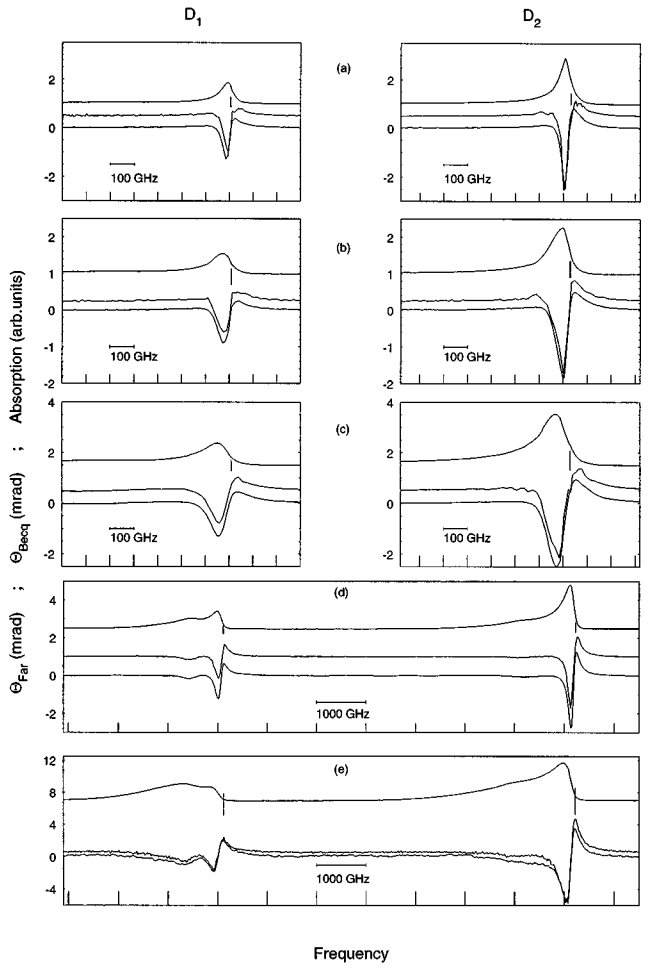

FIG. 1. Survey of experimental absorption spectra and Faraday spectra near the Rb D_1 and D_2 lines. In each box the upper curve gives the absorption spectrum and the lower curve the experimental Faraday spectrum. The middle curve gives the Becquerel prediction of the Faraday spectrum; for clarity it has been given a small vertical offset (the offset is different for the various spectra). The free-atom Rb D_1 and D_2 transition frequencies have been indicated by vertical markers. Note that both the horizontal and vertical scales are different for the various spectra. The cell temperature was slightly different for the various spectra but always within the interval 360–420 K. The Xe density n_{Xe} was (a) 0.84×10^{20} cm^{-3}, (b) 1.2×10^{20} cm^{-3}, (c) 1.7×10^{20} cm^{-3}, (d) 2.9×10^{20} cm^{-3}, and (e) 4.5×10^{20} cm^{-3}; this corresponds to a pressure range $p_{\mathrm{Xe}}=4$–25 atm.

when quasimolecular complications are negligible.

IV. ABSORPTION SPECTRA AND FARADAY SPECTRA

Figure 1 shows the experimental absorption spectra $\kappa(\nu)$ and the Faraday spectra $\theta_{\mathrm{Far}}(\nu)$ for six values of n_{Xe} in the range 0.8–4.5×10^{20} cm^{-3}. At the prevailing temperature (350–400 K) these densities correspond to Xe pressures in the range 4–25 atm. Over this whole range the D_1 and D_2 transitions are well isolated. Both transitions have a very asymmetric line shape: nearly all the transition strength occurs at the red side of the transition frequency of the free Rb atom. This indicates that the impact approximation has lost

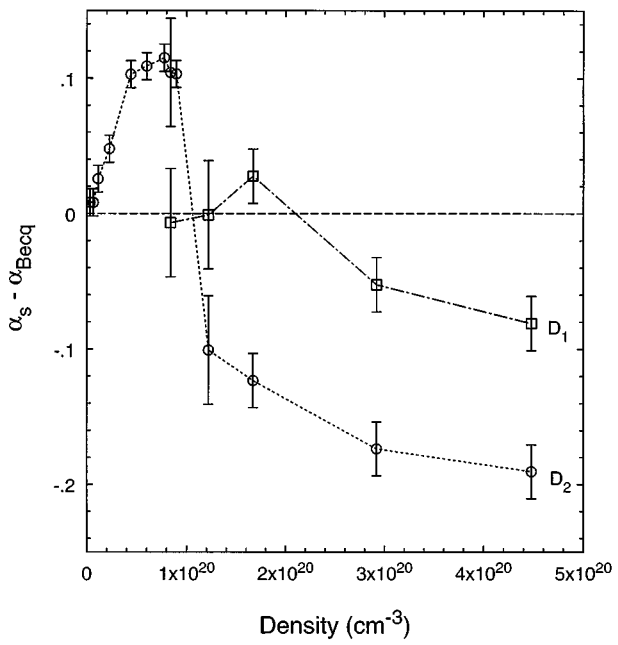

FIG. 2. Deviation of the strength of the Faraday spectrum from the Becquerel prediction. The experimental Faraday spectra in Fig. 1 have been fitted by replacing α_{Becq} in Eq. (1) by a free-scaling parameter α_s. The dashed lines have been drawn to guide the eye.

its validity even in the line core. Both the D_1 and D_2 lines have a satellite in their red wing that becomes visible at higher densities. In particular the D_1 satellite is rather pronounced; for $n_{\mathrm{Xe}}=4.5\times10^{20}$ cm^{-3} [Fig. 1(e)] it is even stronger than the "parent" line. These satellites are due to extrema in the Rb-Xe difference potentials [9,10]. After Kramers-Kronig inversion of the absorption spectra, we calculated the Faraday spectra $\theta_{\mathrm{Becq}}(\nu)$ as predicted by the Becquerel relation [Eq. (1)]; these spectra have been plotted in Fig. 1 together with the measured Faraday spectra $\theta_{\mathrm{Far}}(\nu)$. For clarity, the Becquerel spectra have been given a vertical offset.

A global comparison of $\theta_{\mathrm{Far}}(\nu)$ and $\theta_{\mathrm{Becq}}(\nu)$ in Fig. 1 shows that the *shape* of the Faraday spectra remains surprisingly close to the shape predicted by the Becquerel relation [Eq. (1)]. This is particularly true for the D_1 spectrum; for the D_2 spectrum we observe a slight discrepancy from the Becquerel shape near the satellite in the red wing. This overall preservation of the Becquerel line shape is noteworthy since the corresponding absorption line shapes are extremely non-Lorentzian, i.e., asymmetric. It is not obvious at all that the Becquerel line shape should survive when quasimolecular aspects become important. In fact, in the binary collision regime one finds that the Faraday spectrum can be parametrized as the sum of two terms, one proportional to $dn/d\nu$ and the other to $d\kappa/d\nu$ [5,6,11]; only the former term preserves the Becquerel shape. In that regime we found experimentally [5,6] that the $dn/d\nu$ term was the dominant one ($\sim$75%) for $n_{\mathrm{Xe}}\leqslant1\times10^{20}$ cm^{-3}. A theoretical *calculation* (instead of a parametrization) of this partitioning into two terms has never been performed; this is possible in principle within the context of binary line-broadening theory using the Rb-Xe potentials [12]. Beyond the binary regime ($n_{\mathrm{Xe}}\geqslant1\times10^{20}$ cm^{-3}) a useful theoretical framework does not even exist.

This being said, we will pay no attention to the small discrepancies between the shapes of $\theta_{\mathrm{Far}}(\nu)$ and $\theta_{\mathrm{Becq}}(\nu)$ in Fig. 1 but instead focus on the *amplitudes*. That is to say, we fit the Becquerel expression [Eq. (1)] to our measured Faraday spectra, replacing α_{Becq} by a scaling parameter α_s, which is determined by a least-squares fit. In Fig. 2 we give a plot of $\alpha_s-\alpha_{\mathrm{Becq}}$ versus n_{Xe} for the D_2 and D_1 line. (The error bars correspond to one standard deviation in the least-squares fit.) First we discuss the plot for the D_2 line. This plot shows a surprising amount of structure. The value of $\alpha_s-\alpha_{\mathrm{Becq}}$ increases linearly from zero when the Xe density increases up to $n_{\mathrm{Xe}}\approx0.5\times10^{20}$ cm^{-3}; this is the signature of binary interaction as extensively discussed previously [5,6,13]. The value of $\alpha_s-\alpha_{\mathrm{Becq}}$ reaches a plateau of $\sim+0.1$ for $n_{\mathrm{Xe}}=0.5$–1.0×10^{20} cm^{-3} (this was already noted previously [6]) and then drops suddenly, in a narrow density range around $n_{\mathrm{Xe}}=1\times10^{20}$ cm^{-3}, to ~-0.1. Subsequently, the value of $\alpha_s-\alpha_{\mathrm{Becq}}$ decreases further, but much more slowly, to ~-0.16 at $n_{\mathrm{Xe}}=4.5\times10^{20}$ cm^{-3}.

We interpret this sudden change in the initially linear dependence of $\alpha_s-\alpha_{\mathrm{Becq}}$ versus n_{Xe} as evidence of multiperturber effects on the magnetic precession of the Rb electron in the $^2P_{3/2}$ state. The sudden onset of these effects is surprising. In comparison, we find a more gentle dependence of $\alpha_s-\alpha_{\mathrm{Becq}}$ versus n_{Xe} for the D_1 transition (Fig. 2). In this case the Faraday spectrum has approximately the Becquerel amplitude ($\alpha_s-\alpha_{\mathrm{Becq}}\approx0$) for n_{Xe} up to $\approx2\times10^{20}$ cm^{-3}. For higher values of n_{Xe} we observe a small decrease. This different behavior of the D_1 and D_2 transitions is in line with the fact that in the binary regime Becquerel violation can only occur for the D_2 transition; it is forbidden for the D_1 transition because of the spherical symmetry of both the lower and the upper quasimolecular state [5,6].

V. CONCLUDING REMARKS

We have observed a sudden onset of multiperturber effects in the Faraday spectrum of the Rb D_2 line at Xe densities such that $N_{\mathrm{Xe}}R_W^3\sim1$. We note that a magneto-optical spectrum is apparently a sensitive discriminant for multiperturber effects. Since proper theory is lacking we will restrict ourselves in the following to some speculative comments.

For the D_2 line the initial increase of the Faraday effect (for $n_{\mathrm{Xe}}\leqslant1\times10^{20}$ cm^{-3}) can probably be seen as a consequence of angular momentum recoupling, when proceeding from the atomic (Rb) to the diatomic (Rb-Xe) case. An estimate of the average molecular g factor (i.e., averaged over all Zeeman transitions) gives indeed an α value that is somewhat larger than the atomic value $\alpha_{\mathrm{Becq}}=7/6$.

The decrease of the Faraday effect on the D_2 line for $n_{\mathrm{Xe}}\geqslant1\times10^{20}$ cm^{-3} is probably analogous to the decrease of the Zeeman effect of a bound diatomic molecule when the rotational quantum number J increases [14]. For large J values (either for a bound molecule or for a collision pair) the electronic angular momentum is almost perpendicular to **J** so

that the magnetic moment associated with **J** is very small. This argument does not explain, however, why the presence of *two* (or more) Xe perturbers in the Weisskopf sphere is apparently especially efficient in reducing the magnetic moments. Short-lived Xe_n ($n \geqslant 2$) complexes may play a role in this respect.

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[13] We realize now that in our previous work (Ref. [6]) the linearization of $\alpha_s - \alpha_{Becq}$ was extended to data points that were already somewhat affected by multiperturber effects. A proper fit to a binary theory requires these data points to be omitted. This affects the value of δ'_{Xe} as reported in Ref. [6]: $\delta'_{Xe} = 1.82 \times 10^{-21}$ cm^3 instead of 1.41×10^{-21} cm^3. The values of γ'_{Xe}, δ'_{He}, and γ'_{He} are not affected.
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