Ultrahigh resolution spectroscopy using frequency-stabilized lasers

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Progress in frequency stabilization of dye lasers has encouraged us to undertake serious experimental investigation of those factors that control the ultimate spectroscopic resolution limits. Of course the Doppler broadening due to thermal motion can be eliminated by use of Doppler free techniques such as saturation and two-photon spectroscopy. However, that thermal motion still limits the coherent interaction time through the finite interaction time with bounded laser beams, and ultimately controls the line spectral profile via relativistic effects (see below). Ramsey's method of separated interaction regions[1,2] can provide a major increase of the effective coherent interaction time, especially in connection with collimated atomic beams. For example, in our experiments[3] with the attractive two photon absorption line in Bi [$(6p)^3\ {}^4S_{3/2} \rightarrow (6p)^3\ {}^2P_{3/2}$ at 603.05 nm, $\tau \simeq 4$ ms], a linewidth $\leqslant 1$ kHz would be in principle obtainable for 50 cm separation of the two laser interaction zones. Further, light shifts are strongly reduced ("diluted") since most of the coherent phase difference between laboratory and atomic clocks is evolved while the atom is actually in the dark space between the laser zones. A similar reduction of power broadening occurs in the case of saturation Ramsey fringes. Chebo[3] tayev has also discussed an important angular "multiplex" advantage of the separtaed fields technique in saturation spectroscopy.[4] Based on their potentially-narrow spectral widths and feasible laser wavelengths, attractive two photon transitions include — besides the Bi line — a line in Ag ($4d^{10}5s^2S_1/_2 \rightarrow 4d^9 5s^2\ {}^2D_5/_2$, 661.3 nm, $\tau \simeq 0.25$ s), in Hg^+ ($5d^{10}6s^2S_1/_2 \rightarrow 5d^9 6s^2\ {}^2D_5/_2$, 563.2 nm, $\tau \simeq 0.11$ s), and the lowest ${}^1S \rightarrow {}^1D$ transtitions in the heaviest alkaline earths. To become acquainted with the Ramsey two-zone techniques we have studied two photon Doppler-free transitions to Rydberg states in a Rb atomic beam.[5] The advantage of such transitions is the efficient detection of the excited atomic beam species by field ionization. However, for our purposes, alkali Rydberg atom problems included large ac and dc polarizabilities, copious magnetic and hyperfine structure, and limited level lifetimes ($\sim 25\ \mu s$ at $n^* = 30$). Ramsey fringes of high contrast and width down to ~ 15 kHz were obtained experimentally with 4.2 mm waist separation. Ultimately the dispersion of atomic beam velocities provides the main broadening mechanism via the relativistic time dilation (second-order Doppler effect). Analysis[6] shows that dramatic fringe shape changes occur at high resolution ($\sim$ 1 kHz) consistent with unpublished experimental results by R. L. Barger on the Ca intercombination line at 657.3 nm using 25 cm waist separations. Future work will focus on monovelocity atomic beams (via acceleration or supersonic expansion), or on radiatively cooled samples in electromagnetic ion traps, or perhaps on radiatively-cooled atomic beams, or on longitudinal interaction.

A new laser frequency staiblization concept promises dramatic improvement in frequency stability and linewidths. In preliminary experiments a jet stream dye laser using such techniques was shown to be phase-stable for periods exceeding 10 ms, and so to have a spectral width well below 100 Hz.

References

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稳频激光超高分辨光谱学

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稳频染料激光器的进展鼓舞我们对影响光谱分辨率极限的那些因素进行认真的实验研究。诚然，因热运动引起的多普勒加宽可以用无多普勒技术，例如，饱和吸收和双光子光谱术加以消除。然而，由于通过有界激光束的有限作用时间，热运动仍然限制了相干作用时间，并且最终通过相对论效应（见下文）而控制谱线轮廓。冉赛的分离作用区的方法能够较多地增加有效相干作用时间，特别在准直原子束的情况下更如此。例如，在我们的实验中用603.05毫微米，$\tau \simeq 4$毫秒的Bi $(6p)^3\,{}^4S_{3/2} \to (6p)^3\,{}^2P_{3/2}$相吸的双光子吸收线，当两激光互作用区相距50厘米时原则上可获得$\leqslant 1$千赫的线宽。再者，光频移动也大大地减弱了（“稀释”了），这是因为当原子实际上处在两激光作用区之间的暗空间时，大部分的相干位相差是在实验室时钟与原子钟之间形成的。在饱和的冉赛条纹情况下，功率加宽也出现类似的减弱。戚拜泰耶夫（Chbotayev）曾经讨论过在饱和光谱学中分离光场技术的一个重要的角“多重性”的优点。基于他们的潜在窄线宽和适宜的激光波长，相吸的双光子跃迁——除掉Bi线以外——包括银的一条谱线（$4d^{10}5s^2S_{1/2} \to 4d^9 5s^2\,{}^2D_5/_2$，661.3毫微米，$\tau \simeq 0.25$秒），汞 Hg^+ 的一条谱线（$5d^{10}6s^2S^1/^2 \to 5d^9 6s^{22}D_{5/2}$，563.2毫微米，$\tau \simeq 0.11$秒），以及在最重的碱土族中的最低的 ${}^1S \to {}^1D$跃迁。为了了解冉赛的二个区域的技术，我们研究了在Rb原子束中跃到里德伯态的双光子无多普勒跃迁。这些跃迁的优点是可以用场致电离对激发的原子束样品作有效的探测。然而，就我们的目的而言，碱金属里德伯原子的问题还包含大的交流和直流极化率，丰富的磁结构和超精细结构，以及有限的能级寿命（在$n^*=30$时$\sim$25微秒）。在腰部间隔为4.2毫米的情况下，我们由实验得到了宽度低到$\sim$15千赫的高反差冉赛条纹。最终说来，原子束速度的分散通过相对论时间膨胀（二级多普勒效应）提供了主要的加宽机理。分析表明在高分辨（$\sim$1千赫）情况下出现奇特的条纹形状的改变，这与R. L. 拜格用25厘米腰部间隔在657.3毫微米的Ca内组合谱线的未发表的实验结果是一致的。深入的研究将集中在单一速度的原子束（通过加速或超声膨胀），或者电磁离子俘获中的辐射致冷的样品，辐射致冷的原子束或纵向相互作用上。

一种新的激光稳频概念可望大大提高频率稳定性及线宽。在初期的实验中，采用这一技术的喷射染料激光器表明位相稳定时间可大于10毫秒，因而其谱线宽度远小于100赫。